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Synthesis and crystal structure of some novel phases involving metal-metal bonding

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Synthesis and crystal structure of some novel phases involving metal-metal bonding

by

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INTRODUCTION

Much attention in recent years has been focused on compounds involving metal-metal bonding. While very few examples were known only a few years ago, it now appears that catenation is an important facet of the chemistry of the metallic elements.[1] In approximately the last decade it has been demonstrated that many metals, particularly the heavier ones in lower than normal oxidation states, show a propensity toward forming homonuclear metal to metal bonds. The occurrence of metal-metal bonds in compounds of the transition metals have been reviewed by several authors.[2-5] Recently, Corbett has reviewed the homopolyatomic ions known for the post-transition elements,[6a] and Gillespie and Passmore have presented a general review of the homopolyatomic cations of the elements.[6b] Among the reasons for neglect of this field of chemistry until recently have been the nonexistence of good solvent systems for metallic systems, the range of composition that many such compounds exhibit, and the lack of a theory of valence to adequately describe such compounds.[7] Cotton has emphasized that the surest indication of the existence of a metal-metal interaction is provided by the determination of the molecular structure.[8] In this research X-ray crystallography was chosen as the method of structural elucidation.

The reduction of several of the rare earth metal halides by the respective metal has resulted in several unexpected and unusual compounds.[9] The preparation and subsequent determination of the structure of gadolinium sesquichloride[10,11] represents a rewarding investigation in this area. The overriding feature of the structure is the occurrence of chains of gadolinium atoms involving elongated octahedra sharing edges running parallel to the fiber axis. The chains dictate the structural arrangement and are separated by sheaths of chlorine atoms all of which occupy faces of metal atom triangles.

Research techniques developed in the study of the rare earth-rare earth metal halide systems have proven applicable to the investigation of other systems of experimental interest. A scarcity of information concerning the lower halides of zirconium and hafnium led to the study of the reduction characteristics of halides of the two elements by Struss and Corbett.[12,13] The reaction of gaseous $HfCl_4$ from a reservoir at 450-550° with a large area of hafnium foil at 610-650° in a sealed tantalum tube gives a very small amount of metallic product $HfCl_{1+x}$ ($0<x<.2$). The analogous reaction for $ZrCl_4$ at 600° produces $ZrCl_3$ and a small amount of $ZrCl_{1+x}$ mixed with $ZrCl_2$. Equilibration of either metallic product with the respective metal at 625 to 650° in a tantalum container gives the pure monochloride. While the reduction to zirconium monochloride is more facile, the two monochlorides are apparently

isostructural and metallic conductors.[13] A previous preparation of zirconium(III) chloride, bromide and iodide resulted in better yields than for the corresponding hafnium(III) halides.[14] Apparently the facility of the reduction of zirconium halides relative to those of hafnium is one of the few differences in the chemistry of the two elements. Larsen has proposed a separation scheme involving the reduction of zirconium tetrahalides containing hafnium in natural abundance to the appropriate zirconium trihalide by either metallic zirconium or aluminum in molten 76 mole percent aluminum trihalide at 260-310°.[15] Under the same time and temperature conditions, the respective hafnium tetrahalide remains substantially unreduced and can be separated along with the aluminum halide solvent by sublimation from the nonvolatile zirconium trihalide. It remains to be seen if this approach is practicable, but the separation of zirconium and hafnium is of commercial significance as the disparate thermal neutron capture cross sections of the two elements enjoins the use of zirconium for cladding nuclear fuel and hafnium for control rods in nuclear reactors.[16]

The purity of several of the reported trihalide phases is questionable because of the use of powdered zirconium metal as the reducing agent or because of side reactions with silica containers.[14,17,18] Newham and Watts reported the preparation of pure anhydrous zirconium trichloride, tribromide and

triiodide from passage of the appropriate tetrahalide vapor mixed with hydrogen through a glow discharge.[19] Afterwards the product was freed from absorbed or unreduced contaminants by heating in vacuo. Struss and Corbett carried out their reactions between gaseous tetrahalide and the appropriate metal foil in sealed tantalum containers.[12,13] Thus, side reactions with the container and cessation of the reaction by blockage of the surface of the metal particles were obviated. Zirconium dihalides of uncertain purity have been obtained from the disproportionation of the trihalides.[20,21] Heating zirconium trichloride and metallic zirconium in a quartz tube lined with platinum foil resulted in zirconium dichloride of 95-99% purity.[22] The homogeneity range of zirconium(II) chloride is presently being investigated in this laboratory.[23]

The patent literature contains a reference to a product called "Zirklor"[24] made by electrolytic reduction of a $SrCl_2$-NaCl-$ZrCl_4$ melt (63:34:3) onto graphite. Its color, softness, graphitic character and analysis correspond to the properties of ZrCl, but the powder pattern data given in the patent are in poor agreement with the data reported by Struss and Corbett.[13] Surprisingly, a sample of the material "Zirklor" was found to have substantially the same pattern as did ZrCl.[13] A report of the synthesis of ZrCl and its powder pattern by Troyanov and T'sirelnikov[25] agrees generally with that of Struss and Corbett with the exception of the omission of the lowest angle line in the powder pattern by the former

authors. A subsequent crystal structure determination for ZrCl by Troyanov[26] is poorly refined and in error. The preparation of a suitable single crystal of ZrCl will be described in Chapter II of this thesis and the structure determination of ZrCl will be described in Chapter III.

Several years ago the X-ray investigation of the product of the reduction of molten bismuth chloride by bismuth metal revealed that the phase corresponded to the stoichiometry $BiCl_{1.167}$.[27] The structure determination resulted in the discovery of the homopolyatomic cation Bi_9^{5+} associated with the large complex anions $[BiCl_5]^{2-}$ and $[Bi_2Cl_8]^{2-}$ in the ratio 2:4:1. When the reduction is carried out in the aluminum chloride solvent system, two compounds $Bi_5^{3+}(AlCl_4^-)_3$ and $(Bi^+AlCl_4^-)_n$ crystallize following reduction of the composition $BiCl_3 \cdot AlCl_3$ by a stoichiometric or excess quantity of bismuth, respectively.[28] Attempts to obtain a single crystal of either compound suitable for an X-ray structure determination were thwarted by the tendency to supercool and produce twinned crystals.[29] During the course of this research several attempts were made to prepare the compound $Bi_5^{3+}(AlBr_4^-)_3$ and isolate a single crystal suitable for a structural determination.

Over forty years ago, Zintl reported potentiometric evidence for solutions of polyatomic anions of antimony, bismuth, lead, tin and arsenic, among others, after reduction of a salt of the respective heavy metal with sodium metal in

liquid ammonia.[30-33] The compounds indicated for Group IV and V elements are listed in Table 1. These unusual phases were

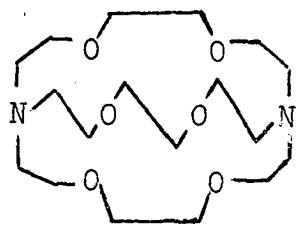
Table 1. Zintl phases

Na_3As	Na_4Sn_9	Na_4Pb_7
Na_3As_3	Na_3Sb	Na_4Pb_9
Na_3As_5	Na_3Sb_3	Na_3Bi
Na_3As_7	Na_3Sb_7	Na_3Bi_3

not further characterized because it was not possible to isolate them from the ammoniacal solutions in crystalline form. It is significant that two of the reported products, Na_4Sn_9 and Na_4Pb_9, contain anions that are isoelectronic and presumedly isostructural with the Bi_9^{5+} cation in $BiCl_{1.167}$.[6] Attempts to prepare solid phases containing these anions have been frustrated by their tendency to form the known and more stable intermetallic phase involving the heavy metal and the sodium counter cation. Some effort has been made to generate such species in molten salts, and the cryogenic evidence for Sb_3^{3-} in NaI is encouraging.[34] Exploratory work in this laboratory seeking to isolate stable cluster anions by varying the conditions, solvent and cation from those used by Zintl have been unsuccessful.[35] However, Kummer and Diehl have

recently reported isolation of a crystalline compound Na_4Sn_9·6-8 ethylenediamine.[36] They present Mössbauer data, magnetic data and preliminary X-ray data, but they have yet to report a complete X-ray structure determination.

Recently, Dye has reported the preparation and crystal structure of a compound $Na_2C_{18}H_{36}N_2O_6$ which contains a cryptated sodium cation and a sodium anion.[37,38] Cryptates are a new type of complexing agent that wrap themselves around metal ions in solution thereby effectively hiding the metal ion from the solvent.[39] The compound $C_{18}H_{36}N_2O_6$[I] is one of



I

a series of polyoxamacrobicycles synthesized by Lehn[40] consisting of a bicyclic ring system with two nitrogen bridgeheads and a defined number of ether-oxygen atoms in the bridges. The systematic designation is 4,7,13,16,21,24-hexaoxa-1,10-diazabicyclo[8.8.8]hexacosane. While the shorter designation 2,2,2-crypt has the feature of specifying the number of ether oxygens in each bridge, in many cases it will be convenient to simply use the abbreviation C. Dye exploited 2,2,2-crypt to encapsulate the sodium cation in solutions of sodium in ethylamine which ultimately allowed

isolation of solid NaC^+Na^-.[37] One sodium cation was found to occupy the central cavity of the macrobicycle[I] as has been found for all of its complexes with alkali, alkaline earth and other metal ions so far studied.[41] The sodium anion was found to be located outside of the cryptated cation at a large distance from all other atoms.[38] By comparison with the known crystal structure of (cryptated sodium) iodide[42] the outside sodium is similar to the iodide ion in its placement, but distance comparisons suggest it is somewhat larger than the iodide ion.[38]

Don Merryman and Frank Armatis have been involved in a series of synthetic attempts to prepare stable compounds containing homopolyatomic anions.[43] Typically, 18-crown-6 ether or the 2,2,2-crypt[I] was allowed to react with sodium alloys of heavy metals in amines to stabilize a salt containing the cryptated sodium cation and a presumed polyatomic anion. In the case of a 1:1 alloy of sodium and antimony, reaction with 2,2,2-crypt in ethylenediamine lead to the isolation of a dark brown crystalline solid on evaporation of the solvent. The X-ray structure determination reported here confirms that the compound involves three cryptated sodium cations and a Sb_7^{3-} cluster anion. The synthesis of $(NaC^+)_3Sb_7^{3-}$ will be described in Chapter II of this thesis. The details of the structure determination will be given in Chapter IV.

During the course of this work the goal of synthesizing a single crystal of zirconium monochloride suitable for a structure determination and solving the crystal structure has been realized. Now that the structure is known, the physical properties can be correlated with it. In fact, the layer structure determined for zirconium monochloride explains why it was so difficult to find a suitable single crystal. The synthetic work on the Bi-$BiBr_3$-$AlBr_3$ system did not produce such satisfactory results. While apparently crystalline material was obtained from two preparations and X-ray data sets were recorded for two different crystals, it has not been possible to solve the structure. Reasonable positions for the tetrabromoaluminate groups could be chosen by inspection of the Patterson map, but the positions of the bismuth atoms were more elusive. One interpretation of the problem involves packing of the Bi_5^{3+} units in alternative orientations. Despite the disappointment of not having solved the structure, the structure solution of $(NaC^+)_3Sb_7^{3-}$ has provided the opportunity to extend the idea of catenation of metal atoms to anions and to demonstrate the first example of a stable solid containing a well-characterized homopolyatomic cluster anion for a metallic element.

EXPERIMENTAL PROCEDURE

Preparation and Handling of Compounds

Preparation of zirconium monochloride

Struss and Corbett reported the formation of stable zirconium monochloride from the reaction of a partially reduced sample of zirconium chloride with zirconium metal at 625° in a closed tantalum container.[13] The purpose of this research was to prepare a single crystal of ZrCl suitable for a structure determination. While the preparation of ZrCl was quite straightforward, the first several reactions resulted in polycrystalline products. The metal strips used had been cold-rolled to a 15-20 mil fold from crystal bar zirconium containing less than 0.05% hafnium. Typically two strips of foil about 8 cm long were reacted with 0.05 g of zirconium tetrachloride in a closed tantalum container. The tetrachloride had been prepared by treating the metal with HCl (Precision Gas Products) at an initial temperature of 300° and increasing to 500° followed by vacuum sublimation through a course frit.[13] In order to determine the optimum temperature conditions, several preparations were carried out at various temperatures between 650 and 900°. The use of a 700-900° temperature gradient gave somewhat better results, but the reaction period of one week was apparently too short as the crystals were too small. A subsequent preparation was

carried out under similar conditions for 30 days. The half of a metal strip that had been at the cool end of the reaction tube was covered with gleaming black zirconium monochloride platelets while a dull grey powder clung to the other half of the strip. The powder pattern of the ZrCl platelets compared favorably with that of Struss and Corbett,[13] and the powder pattern of the grey powder established it as zirconium metal. It is reasonable that ZrCl would disproportionate to $ZrCl_4$ and the metal at 900°. Several platelets of ZrCl from this preparation were mounted and exposed to X-rays. Most were polycrystalline, but a few gave streaked spots. The layer line separation in these photographs gave a spindle axis of 3.42Å. The idea that the streaking indicated a disordering problem prompted the construction of a device capable of producing a continuously increasing potential that could be attached to the thermocouple input of a Brown temperature controller to enable the reaction tube to be cooled very slowly. The effect of electropolishing the metal was also contemplated, and a series of preparations was carried out to assess the merit of these proposals. The best crystals obtained from this series of reactions were grown on electropolished metal in a 700-900° temperature gradient. Evidently electropolishing the metal does help to prevent nucleation, and electropolishing is the method of choice for cleaning zirconium because the high affinity of zirconium for oxygen

precludes cleaning it in oxidizing acid solutions or by induction heating. Finally, a preparation was undertaken in which electropolished strips of zirconium metal were heated with a small amount of zirconium tetrachloride in a sealed tantalum container in a 600-800° gradient for 24 days. The furnace was then cooled at an initial rate of 1.25° day^{-1} for four days. At this point the rate was increased to 5° day^{-1}. The scheme of slowly cooling the furnace was thwarted the next day by a power failure. Nonetheless, when the tube was taken into the dry box and cut open, bright, well-formed hexagonal platelets were found clinging to the center portions of the metal strips. No reaction had occurred at the cool end of the tube. As before, powdered zirconium metal was found on the surface of the strips at the hot end of the tube. Of the dozen crystals that were mounted, two gave sufficiently promising oscillation patterns that they were considered good enough to align. Oscillation photographs established that the spindle axis was 3.42Å as before. However, the zero-level and first-level Weissenberg photographs of these two crystals were now of sufficient quality to allow construction of the reciprocal lattice. Plotting the spots from the two films on polar graph paper resulted in a primitive monoclinic cell having a=9.19, b=3.42, c=5.92Å and β=103°. Unfortunately, by the time that priority for the diffractometer could be established, neither crystal diffracted

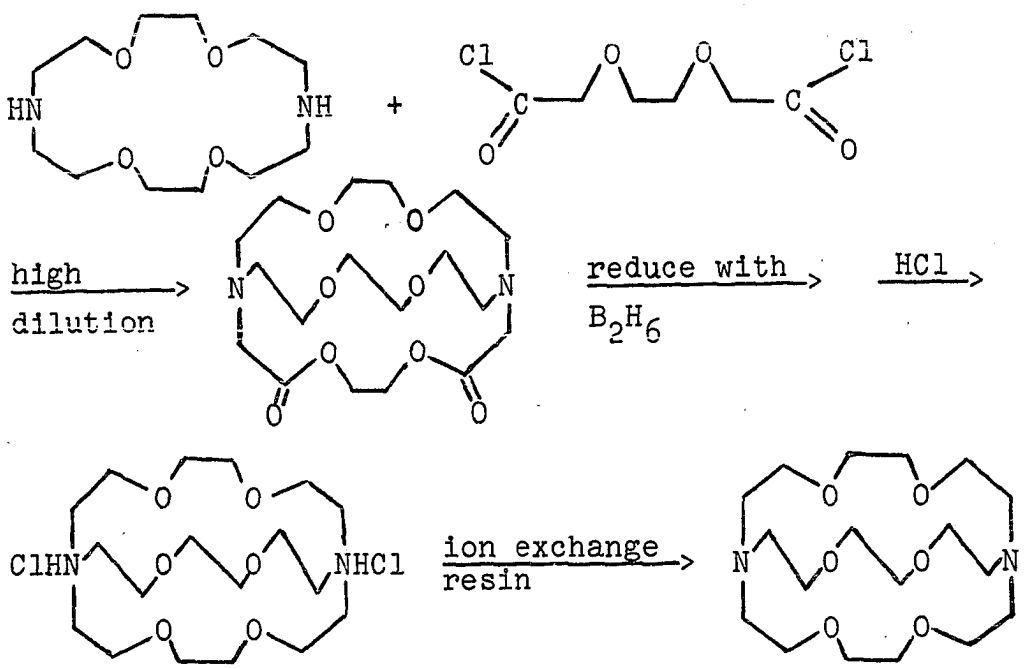
X-radiation. Since a search of the reaction product from which these two crystals had originally been obtained failed to produce a suitable crystal, the reaction was repeated using the same conditions. This reaction was apparently defeated by power failures during both the equilibration and cooling periods. After an appeal for an emergency power circuit had been heeded by the lab electricians, the reaction was repeated yet again. Following two sessions of mounting crystals from this reaction, capillary No. 215 was confirmed to contain a single crystal of zirconium monochloride.

Preparation of (4,7,13,16,21,24-hexaoxa-1,10-diazabicyclo-[8.8.8]hexacosane) sodium heptantimonide(-3)

Preliminary X-ray investigations were carried out on several phases expected to contain polyanions. In their synthetic work, Don Merryman and Frank Armatis employed the bicyclic amine 2,2,2-crypt or 18-crown-6 ether with sodium alloys of bismuth, lead and antimony in a solvent such as ethylenediamine or liquid ammonia to obtain a solid presumably containing a polyanion.[43] The first such solid investigated resulted from the reaction of NaSb and 2,2,2-crypt in ethylene-diamine. Microscopic examination of the resultant solid product revealed dark brown crystals possessing regular external morphology in the form of needles on the order of tenths of millimeters in diameter and a millimeter in length. The first crystals that were mounted in capillaries in the

dry box were picked up with a wand greased with Vaseline. Two of six crystals mounted initially exhibited a diffraction pattern, but after several days they no longer diffracted X-rays. Crystals from a second preparation of this compound were mounted on another occasion using silicone lubricant rather than Vaseline. Several of these crystals gave good diffraction patterns. The reaction of $NaSb_3$ with 2,2,2-crypt in ethylenediamine produced a solid of similar morphology. Diffraction photographs obtained from these crystals had the same pattern as those obtained from the material prepared from the 1:1 alloy. Powder patterns of the two solids confirmed that they were the same compound. Several other compounds were examined with less success. A grey solid made by mixing NaBi and 2,2,2-crypt in ethylenediamine occurred as diamond shaped crystals adhering to the walls of the reaction tube. Once the tube was cracked open in the dry box, it was discovered the crystals had the consistency of Philadelphia cheese. None was successfully mounted in a capillary. A dark brown solid made from Na_4Pb_9 and 18-crown-6 ether occurred as large needles. While these crystals were easily scraped from the tube and mounted, most were found to be amorphous upon exposure to X-rays. A second preparation involving Na_4Pb_9 and 2,2,2-crypt in liquid ammonia gave slightly better formed crystals. However, these too were shown to be amorphous upon X-ray examination.

Since the antimony system seemed the most promising, further effort was concentrated on that system. Good crystals were readily obtained by reacting powdered NaSb with 2,2,2-crypt in ethylenediamine at room temperature for 12 hours.[43] The reaction was carried out in a Y-shaped reaction vessel which could be evacuated. Afterwards, the side arm was immersed in an ice bath and the ethylenediamine solvent was slowly distilled off over a twelve hour period. A deficiency of crypt was used to favor cryptation of sodium ions. The 1:3 alloy which was found to give the same product dissolved more slowly. Letting the solution stand longer or gently heating it to 40-50° was found to be equally satisfactory. The 2,2,2-crypt used in these syntheses was purchased from EM Laboratories, Inc. Its synthesis has been outlined by Truter and Pedersen[39] as follows:



Dye, et al. have published a modification[44] of the original procedure of Lehn.[45]

Mounting of crystals

A dry box designed primarily for mounting crystals was obtained from Blickman, Inc. It differs from the other dry boxes in the group in having a nearly horizontal window to facilitate the use of an external Bausch and Lomb "Stereozoom" microscope with a 7 inch focal length. To avoid the inconvenience of focusing the microscope from outside of the box, crystals to be mounted are placed on a lab jack which can be raised or lowered as needed to bring them into focus. This dry box has been equipped with a hot wire for sealing off capillaries which is operated by a foot switch. The moisture content of the box is monitored with a Beckman electrolytic hygrometer. Like the other dry boxes in the group, entrance is gained through an evacuable port. The box is flushed with dry nitrogen. An internal recirculating system consisting of a squirrel cage fan which pulls the incoming gas stream and the box atmosphere through a Molecular Sieve is capable of keeping the moisture content in the box below 30 ppm under most circumstances.

After crystals were sorted in the dry box, candidates for mounting were picked up on the end of a thin glass stalk tipped with Vaseline (ZrCl) or silicone lubricant ($[NaC^{+}]_3Sb_7^{3-}$) and gently inserted into 0.2 mm i.d. Lindemann

($LiBeBO_3$) glass capillaries. Once the stalk was inserted into the capillary, it was twisted to coat the walls with lubricant so that it could be removed leaving the crystal lodged in the capillary. The end of the capillary was then sealed off using the resistance wire inside the dry box, and the seal-off was checked visually with the microscope for the absence of pin-holes. When enough crystals had been mounted, the capillaries were removed from the dry box and sealed off to a length of about one cm on either side of the crystal using a fine tipped gas-oxygen flame. These shortened capillaries were then held in a brass pin 7 mm long by 3 mm o.d. with Apiezon W. Finally, the pin was placed in a goniometer head which could be attached to either X-ray cameras or the full-circle diffractometer.

X-Ray Diffraction Techniques

Camera techniques

Prior to the collection of intensity data, oscillation photographs were taken on a Weissenberg camera[46] in order to assess the diffraction characteristics of the crystal. If it is desired to proceed with further film work, the crystal is aligned so that the spindle axis is perpendicular to the X-ray beam. It is then possible to take Weissenberg photographs from which the reciprocal lattice can be constructed. The Weissenberg method involves translating the camera back and forth while the crystal oscillates. A screen with a

slit in it allows only one predetermined level of diffraction maxima to expose the film. In this manner the spots of the given diffracted cone are spread out over the film in a series of pinacoids which can be readily indexed. If, for example, the spindle axis corresponds to the b-axis, then the zero-level photograph contains all the h0ℓ reflections, the first level the h1ℓ, the second level the h2ℓ, and so forth. In order to examine the extinctions along the spindle axis it is either necessary to realign the crystal along another axis or to obtain photographs using a precession camera. The precession technique allows undistorted reproduction of the reciprical lattice.[47] For a crystal having the b axis corresponding to the spindle axis, this is accomplished by aligning the crystal so that the hk0 or 0kℓ plane is normal to the X-ray beam. A flat piece of film opposite the beam from the crystal precesses while the crystal is slowly precessed. A screen with a circular slit enables diffraction maxima from a given layer to be recorded on the film. The spindle axis is then rotated by the angle β* in order to record the diffraction maxima from the other plane.

Data collection

Intensity data for the reflections observed within one asymmetric unit of reciprocal space for a given crystal system can be obtained either by estimating intensities of spots on film or by using an automated diffractometer. X-ray data for

both of these compounds were taken at ambient temperature using a four-circle diffractometer interfaced with a PDP-15 computer.[48] The computer is programmed to control the diffractometer such that it is possible to take data from an unaligned crystal.[49] Several photographs are taken at arbitrary values of χ and ϕ. A few spots are chosen on each photograph, and their coordinates become input to the computer. The computer indexes these ten to fifteen reflections based on the fact that the indices must be whole small numbers. It then outputs a tentative unit cell and cell scalars which the user may transform if it is necessary. This unit cell assignment is verified by taking "oscillation" photographs aligned along the three crystallographic axes. Finally, three strong noncoplanar reflections are chosen as standard reflections. The instrument tunes on these reflections prior to undertaking data collection, and it checks these standards periodically while data are being collected. The X-radiation employed is Mo K_α (λ=7.0954Å) obtained using a graphite monochromator. Reflections are measured with a scintillation counter using a step scan technique[50] to obtain integrated intensities.

ZrCl Diffraction data for ZrCl were collected from a shiny black hexagonal platelet measuring 0.14 x 0.11 x 0.02 mm. The dataset was recorded on the basis of a monoclinic unit cell having a=5.95, b=3.43, c=9.09Å and β=102.43°. This

compares with lattice constants a=9.19, b=3.42, c=5.92 and β=103° estimated from film work with a previous crystal that proved to be unsuitable for a complete structure determination. A total of 230 integrated intensities were obtained out of 416 reflections checked in the two octants hkℓ and hk$\bar{\ell}$. Refined unit cell parameters and their standard deviations were obtained by a least squares fit[51] to twice the Ω angles of 16 independent reflections which were tuned by the diffractometer after the dataset had been recorded. Any error in the instrumental zero was eliminated by centering the reflection at +Ω and -Ω. The final cell constants with errors for the last significant digits in parentheses are a=5.943 (6), b=3.419 (3), c=9.087 (18) Å, α=γ=90.00 (0) and β=102.50 (12)°.

$(NaC^{+})_3Sb_7^{3-}$ The needle used for data collection was an approximately regular rectangular prism having dimensions 0.1 x 0.2 x 0.6 mm. The compound crystallizes with monoclinic symmetry: a=23.45, b=13.93, c=25.52Å and β=108.73°. A total of 6717 integrated intensities were measured for reflections hkℓ and hk$\bar{\ell}$ out of 11,263 reflections scanned. Since a thirty per cent decrease in the intensities of the standards occurred during data collection, a dataset scaled linear in time to the initial sum of the intensities of the standards was prepared. Lattice constant refinement in the same manner as for ZrCl resulted in final cell constants a=23.263 (43), b=13.776 (4), c=25.344 (73) Å, α=γ=90.00 (0) and β=108.57 (19)°.

Data reduction

After data collection, a series of steps collectively referred to as data reduction are carried out. Initially the data tape was read, and cards were punched with a record of the indices, theta, chi, phi, background count, total count and net count for each reflection. At this point the raw intensities were corrected for absorption. The extent to which an X-ray is absorbed in passing through a crystal of thickness t is given by[52]

$$I=I_o \exp(-\mu t) .$$

In this expression μ, the linear absorption coefficient, is defined as

$$\mu=\frac{n}{V} \sum_i (\mu_a)_i$$

where μ_a is the atomic absorption coefficient and n is the number of molecules in the unit cell with volume V. The absorption by a given crystal is thus a function of its shape and its orientation. No fully satisfactory method of evaluating the absorption correction for the general case has so far been described. The difficulty in making absorption corrections arises from having to calculate the absorption for the actual path length traveled within a crystal by the incident and diffracted beams for each infinitesimal volume element of the crystal dV and then to integrate these results

over the entire volume of the crystal.[53] While the linear absorption coefficients for ZrCl and $(NaC^{+})_3Sb_7^{3-}$ were both small, 66 and 25, respectively, the dimensions of both crystals were sufficiently different that it seemed appropriate to apply an absorption correction. The program TALABS[54] which was used calculates the absorption coefficient[55]

$$A=\int \exp(-\mu L)dV \cdot$$

The total path length L is equal to the sum of l_1 and l_2 where l_1 is the length of the path of the incident ray from its point of entry into the crystal up to the volume element dV and l_2 is the corresponding length of the path of the beam diffracted in dV. The integration is carried out over the volume V of the crystal. The program bases this calculation on the contribution to the diffracted intensity from a tetrahedron in which the path length of the rays is a linear function of the coordinates of the diffracting element.

After the absorption correction is carried out, each dataset was reduced using the program DATRED. This program divides each integrated intensity by its absorption coefficient, A, and also by a Lorentz and polarization factor[56] given by

$$L_p = \frac{\cos^2 2\theta_m + \cos^2 2\theta}{(1 + \cos^2 2\theta_m)\sin 2\theta} \cdot$$

These corrected intensities are proportional to the square of the structure factor, F_o^2. The estimated error in each intensity measurement is calculated by

$$(\sigma_I)^2 = [TC + BK + (K_T TC)^2 + (K_B BK)^2 + (KI)^2]/A^2$$

where TC is the total count, BK is the background count, I is the net intensity, A is the transmission factor, and K_T, K_B and K are fractional random errors in TC, BK and I, respectively.[57] The arbitrary value 0.03 was assigned to K_T. K_B and K. The estimated standard deviation in each structure factor is based on the finite difference method. The function used is

$$\sigma_F = \left| \frac{I/A + \sigma_I}{L_P} \right|^{\frac{1}{2}} - |F_o|$$

where all terms are as defined above.[57] In the case of ZrCl 203 reflections of the 230 integrated intensities collected were found to be statistically above background by more than $3\sigma_I$. Of the 6717 integrated intensities recorded for $(NaC^+)_3Sb_7^{3-}$, 4662 were more than $3\sigma_I$ above background.

Space group determination

Each reduced dataset was inspected for systematic conditions for reflection. For ZrCl the condition h+k=2n for all reflections hkℓ was observed. This condition requires the cell to be C-centered. Since no other conditions were

noted, the space group choice included C2 No.5, Cm No.8 and C2/m No.12. In the case of $(NaC^{+})_3Sb_7^{3-}$ no conditions were found for hkℓ reflections. The condition k=2n found for 0k0 reflections requires a 2-fold screw axis and the condition h+ℓ=2n for h0ℓ reflections requires an n-glide. These conditions serve to uniquely fix the space group as $P2_1/n$ (a non-standard setting of $P2_1/c$).

THE CRYSTAL STRUCTURE OF ZIRCONIUM MONOCHLORIDE

Structure Determination

Structure solution

The solution of a crystal structure from the intensities of a set of X-ray diffraction maxima depends upon recovering the phase relationships among the maxima. The intensity of scattered radiation is proportional to the absolute value of the square of the amplitude $|F(hk\ell)|^2$. The structure factor $F(hk\ell)$ represents the resultant amplitude of the waves scattered in the direction of the reflection $hk\ell$ by the j atoms in the unit cell:

$$F(hk\ell) = \sum_j f_j \exp[2\pi i(hx_j + ky_j + \ell z_j)].^1$$

The value of the structure factor is determined by the atomic scattering factors f_j, which are a function of the number and distribution of electrons in the atoms and on the scattering angle θ, and by the exponential terms, which depend on the positions of the atoms. Since X-rays are scattered by electrons, solution of a crystal structure is equivalent to finding the electron density. The electron density is given

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[1] A more complete discussion can be found in any standard crystallographic text such as Buerger.[46]

by the Fourier transform of the structure factors:

$$\rho\ (x,y,z) = \frac{1}{V} \sum_h \sum_k \sum_\ell F(hk\ell)\ \exp[-2\pi i(hx + ky + \ell z)].$$

As stated earlier, it is necessary to devise a way to determine the phases of the structure factors in order to solve the crystal structure. No general solution to the phase problem exists. It has been found, however, that structures containing one or a few atoms that are markedly heavier than the remainder can be solved by locating the heavy atoms by methods which do not require a prior knowledge of the phases. The heavy atoms can then provide the principal phasing from which the lighter atom positions can be developed. Patterson pointed out that a Fourier calculation using the phaseless quantities $|F|^2$ results in peaks corresponding to all the interatomic vectors, with magnitudes proportional to the atomic numbers of the atoms involved.[58]

A three dimensional Patterson map was calculated from the 203 observed structure factors for ZrCl using the program ALFF.[59] Interpretation of the Patterson map was very straightforward. Four strong peaks of equal intensity were located at $\frac{3}{4},0,\frac{1}{4}$, $\frac{1}{4},\frac{1}{2},\frac{1}{4}$, $\frac{1}{4},0,\frac{3}{4}$ and $\frac{3}{4},\frac{1}{2},\frac{3}{4}$. Inspection of the interatomic vectors for the general positions of the three possible space groups prompted the choice of C2 No.5 because of its intraset vectors $\pm(2x,0,2z;\ \frac{1}{2}+2x,\frac{1}{2},2z)$. The

peak at $\frac{3}{4},0,\frac{1}{4}$ was assigned as corresponding to a fourfold zirconium position. Three cycles of full matrix least squares refinement on the x and z coordinates of one zirconium at $\frac{3}{8},0,\frac{1}{8}$ using the program ORFLS[60] resulted in an unweighted residual[1] of 0.384. The chlorine position was located from an electron density map calculated using the program ALFF.[59] Other than the zirconium peak at $\frac{3}{8},0,\frac{1}{8}$ and the peak at $\frac{1}{8},0,\frac{3}{8}$ assigned as chlorine, the map was flat. Three cycles of refinement on the x and z coordinates of these two atom positions resulted in unweighted residual of 0.283.

Refinement

Refinement of the structure was carried out using the full matrix least squares program ORFLS.[60] The scattering factors used are those of Hanson et al.[61] with corrections for both real and imaginary parts of anomalous dispersion.[52] The reflections were weighted by σ_F^{-2} to account for the reliability of each measurement. A reduced dataset that had

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[1]The unweighted residual R and the weighted residual R_w used are defined by

$$R = \Sigma\left|\,|F_o| - |F_c|\,\right|/\Sigma|F_o|$$

and $$R_w = [\Sigma_w(|F_o| - |F_c|)^2/\Sigma_w|F_o|^2]^{\frac{1}{2}}.$$

not been corrected for absorption was used in the initial stages of the refinement. A total of five cycles of refinement on the x and z coordinates of the two atom positions resulted in a residual of 0.146. Since the space group does not fix y, the y coordinate of one atom must be held constant. However, it was found to be necessary to constrain the y coordinates of both atoms in order to carry out the refinement using this dataset. The refinement of the x and z positional coordinates and the isotropic temperature factors of the two atoms converged in two cycles with R=0.120 and R_w=0.150. Final positional parameters and isotopic temperature factors are listed in Table 2. The specter of partial occupancy was

Table 2. Atomic parameters[a] from isotropic refinement of ZrCl

	Positional parameters			Isotropic thermal parameters
	x	y	z	B(x10^3)
Zr	0.3780(6)	0.0000(0)	0.1339(4)	893(123)
Cl	0.1100(17)	0.0000(0)	0.3295(10)	995(183)

[a]Estimated standard deviations in parameters from ORFLS[60] are in parentheses.

ruled out after varying the atom multiplier for zirconium. In two cycles of refinement the atom multiplier for zirconium

converged at 0.987(25) with R=0.121 and R_w=0.150. A difference electron density map was flat to ±2 electrons/Å^3. When the refinement was repeated using the absorption-corrected dataset it was possible to vary the y coordinate for the chlorine atom. However, since the same weighted residual of 0.165 for the converged positional and isotropic thermal parameters was obtained as in the case where y for chlorine was constrained to zero, the latter case is the preferable one according to the statistical test of Hamilton.[62]

Conversion of the isotropic temperature factors to anisotropic temperature factors brought even more headaches. It was not possible to vary the anisotropic temperature factors[1] for either atom without their becoming not positive-definite. Refinement of the x and z positional parameters and the components of the anisotropic temperature factors β_{11}, β_{33} and β_{13} while damping the shifts in β_{22} and holding β_{23} and β_{12} identically zero using the original reduced dataset resulted in the parameters shown in Table 3 and R=0.081, R_w=0.109. The observed and calculated structure factors are listed in Table 4. In the refinement using the dataset that

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[1]The anisotropic temperature factors have the general form $\exp-(h^2\beta_{11} + k^2\beta_{22} + \ell^2\beta_{33} + 2hk\beta_{12} + 2h\ell\beta_{13} + 2k\ell\beta_{23})$.

Table 3. Atomic parameters[a] from anisotropic refinement of ZrCl

	Positional parameters			Anisotropic thermal parameters ($\times 10^4$)			
	x	y	z	β_{11}	β_{22}	β_{33}	β_{13}
Zr	0.3779(5)	0.0000(0)	0.1339(3)	58(10)	8[b]	68(5)	11(5)
Cl	0.1104(14)	0.0000(0)	0.3306(9)	72(23)	5	69(10)	20(12)

[a]Estimated standard deviations in parameters from ORFLS[60] are in parentheses.

[b]Errors indeterminant since shifts were damped.

had been corrected for absorption, the anisotropic temperature factors again became not positive-definite. For the case where the y coordinate for chlorine as well as β_{12} and β_{23} were constrained at zero and where the shifts in β_{22} were damped, residuals R=0.081 and R_w=0.111 were obtained.

Improving the refinement

Inquiry concerning the peculiarities encountered in the refinement of the dataset collected from this crystal disestablished their uniqueness. Apparently, the program controlling the data-taking procedure of the diffractometer contained a statistical error for the larger reflections which are normally counted for a shorter period of time than the

Table 4. Observed and calculated structure factors[a] for ZrCl

L = 0			
H	K	FO	FC
2	0	60	71
4	0	583	606
6	0	111	103
1	1	237	230
3	1	257	357
5	1	153	140
0	2	1136	890
4	2	486	505
6	2	75	94
1	3	170	161
3	3	179	166
0	4	570	459

L = 1			
H	K	FO	FC
0	0	353	316
2	0	209	206
4	0	221	235
6	0	135	128
1	1	812	817
3	1	174	165
5	1	428	439
0	2	342	363
2	2	180	175
4	2	207	201
1	3	536	525
3	3	102	112
0	4	177	151

L = 2			
H	K	FO	FC
0	0	258	232
2	0	695	658
4	0	210	197
6	0	351	329
1	1	291	292
3	1	211	209
5	1	221	211
0	2	184	168
2	2	502	534
4	2	164	172
1	3	223	206
3	3	147	135
0	4	115	105

L = 3			
H	K	FO	FC
0	0	247	217
2	0	314	363
4	0	190	182
6	0	211	192
1	1	261	261
3	1	506	450
5	1	185	193
0	2	191	208
2	2	230	252
4	2	147	157
1	3	176	178
3	3	335	348

L = 4			
H	K	FO	FC
0	0	667	553
2	0	267	304
4	0	338	315
1	1	238	372
3	1	294	255
5	1	176	172
0	2	471	452
2	2	218	227
4	2	264	280
1	3	138	159
3	3	202	203

L = 5			
H	K	FO	FC
0	0	381	382
2	0	224	232
4	0	221	215
1	1	357	384
3	1	218	226
5	1	200	196
0	2	269	254
2	2	169	198
4	2	189	194
1	3	266	283

L = 6			
H	K	FO	FC
0	0	313	275
2	0	228	254
4	0	208	186
1	1	241	247
3	1	228	216
0	2	228	227
2	2	181	222
1	3	177	196

L = 7			
H	K	FO	FC
0	0	296	244
2	0	177	196
4	0	227	208
1	1	210	211
3	1	163	151
0	2	243	212
2	2	157	177

L = 8			
H	K	FO	FC
0	0	206	167
2	0	147	163
1	1	231	229
3	1	135	132
0	2	158	147
2	2	120	148

L = 9			
H	K	FO	FC
0	0	149	143
2	0	202	214
1	1	98	101
0	2	146	130

L = 10			
H	K	FO	FC
0	0	123	117

L = -1			
H	K	FO	FC
2	0	237	229
4	0	177	206
6	0	156	155
1	1	55	72
3	1	780	866
0	2	374	363
2	2	198	213
4	2	150	154
6	2	135	139
3	3	492	493
0	4	199	151

L = -2			
H	K	FC	FC
0	0	238	232
2	0	834	804
6	0	460	454
1	1	211	206
3	1	271	349
5	1	155	151
0	2	200	169
2	2	589	619
4	2	63	42
6	2	407	412
1	3	162	147
3	3	152	162
0	4	119	105

L = -3			
H	K	FO	FC
0	0	234	217
2	0	292	284
4	0	159	166
6	0	144	148
1	1	748	655
3	1	169	164
5	1	485	481
0	2	199	212
2	2	241	235
4	2	137	140
6	2	133	133
1	3	478	460
3	3	86	111

L = -4			
H	K	FO	FC
0	0	627	553
2	0	274	256
4	0	453	501
6	0	97	101
1	1	334	359
3	1	211	200
5	1	180	185
0	2	484	454
2	2	213	200
4	2	396	433
6	2	83	92
1	3	226	217
3	3	121	129

L = -5			
H	K	FO	FC
0	0	363	382
2	0	243	358
4	0	222	231
6	0	115	117
1	1	274	301
3	1	494	432
5	1	154	162
0	2	272	255
2	2	194	177
4	2	203	202
1	3	189	201
3	3	309	334

L = -6			
H	K	FO	FC
0	0	295	275
2	0	388	369
4	0	194	211
6	0	257	301
1	1	220	225
3	1	272	240
5	1	137	140
0	2	233	228
2	2	321	307
4	2	176	188
1	3	165	171
3	3	179	192

L = -7			
H	K	FO	FC
0	0	277	244
2	0	251	257
4	0	184	177
6	0	187	171
1	1	213	246
3	1	226	215
5	1	255	249
0	2	218	213
2	2	207	208
4	2	159	157
1	3	161	194

L = -8			
H	K	FO	FC
0	0	181	167
2	0	205	202
4	0	209	197
1	1	171	191
3	1	209	198
5	1	194	171
0	2	136	147
2	2	171	181
4	2	166	177

L = -9			
H	K	FC	FC
0	0	158	143
2	0	210	216
4	0	173	158
1	1	147	158
3	1	137	136
0	2	138	130
2	2	192	195

L = -10			
H	K	FO	FC
0	0	131	117
2	0	87	93
4	0	138	131
1	1	179	204
3	1	112	121

[a]Structure factors have been multiplied by 10.

smaller ones and rescaled. When this problem was investigated it was also found that a set of parentheses missing in the program declaring the orientation matrix had the result of equating θ and Ω. A nonuniformity in the steps of the Ω scan was also discovered.[63] While the quantitative effect of these problems is difficult to assess precisely, it is considered fortunate that the refinement of this dataset went as well as it did. Particularly in view of the fact that the counting statistics were in question for the larger reflections it seemed appropriate to reweight the dataset. New weights were assigned to each reflection of the original reduced dataset using the program Omega.[1] All positional and thermal parameters converged after two cycles of refinement using the new weighting scheme. The final residual indices were R=0.080 and R_w=0.087. The parameters from this refinement are given in Table 5. The positional parameters for both zirconium and chlorine are within one standard deviation of those reported in Table 3 for the refinement using the original weighting scheme. However, the errors in these parameters

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[1]Omega is a local program written by C. R. Hubbard which calculates new weights for structure factors after refinement of a structure using ORFLS.[60]

Table 5. Final parameters[a] from the refinement of ZrCl using new weights

	Positional parameters			Anisotropic thermal parameters ($\times 10^4$)			
	x	y	z	β_{11}	β_{22}	β_{33}	β_{13}
Zr	0.3779(2)	0.0000(0)	0.1341(2)	31(7)	3(22)	63(3)	11(3)
Cl	0.1098(7)	0.0000(0)	0.3296(6)	43(13)	21(38)	67(5)	17(6)

[a]Estimated standard deviations in parameters from ORFLS[60] are in parentheses.

are improved by reweighting, as expected. Happily, the thermal parameters converged in this refinement without anomalous behavior on the part of β_{22}. Reweighting the reflections of the dataset that had been corrected for absorption and carrying out one cycle of least squares refinement resulted in the residuals R=0.082 and R_w=0.098.

A projection of the structure along the unique b-axis is presented in Figure 1. An alternate projection on the (001) plane is shown in Figure 2. The thermal ellipsoid plot program ORTEP[64] was used to prepare these drawings.

Interatomic distances and angles

Interatomic distances and angles were calculated using the function and error program ORFFE.[65] The estimated

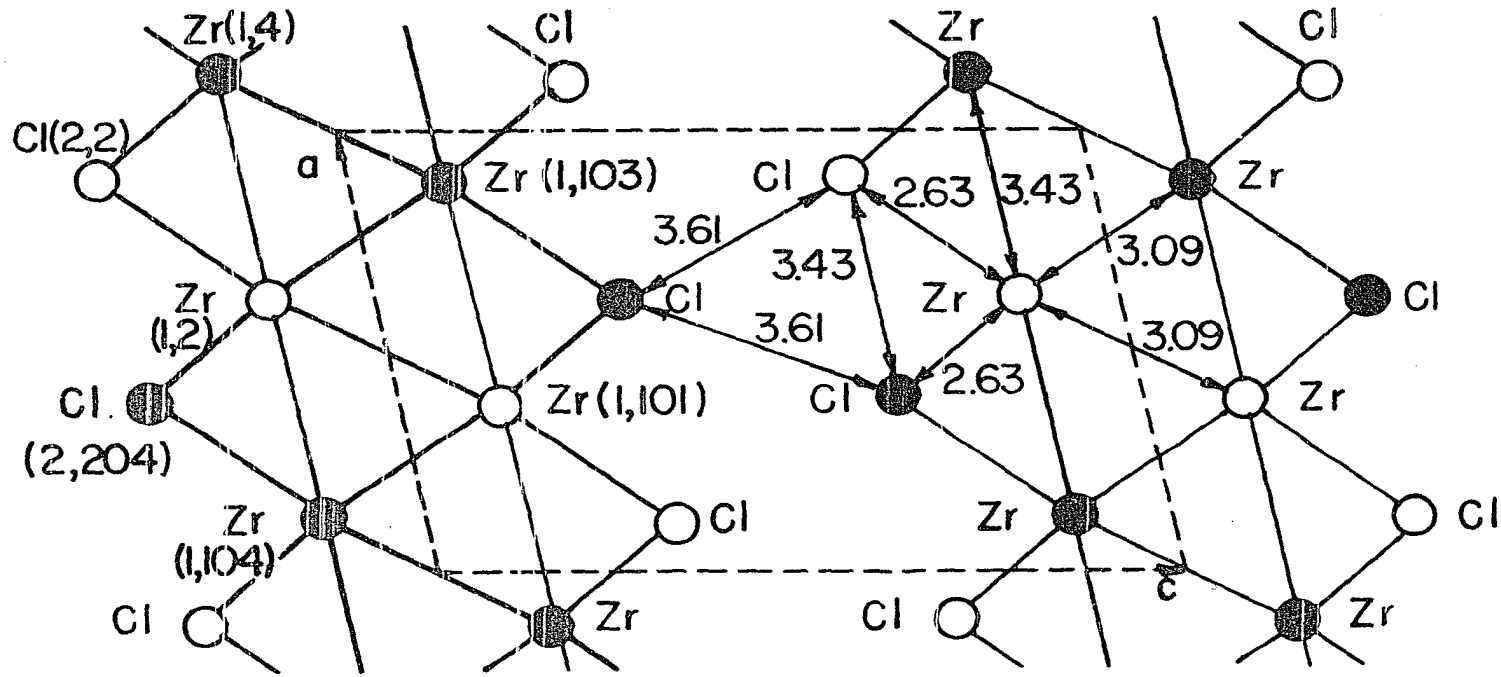
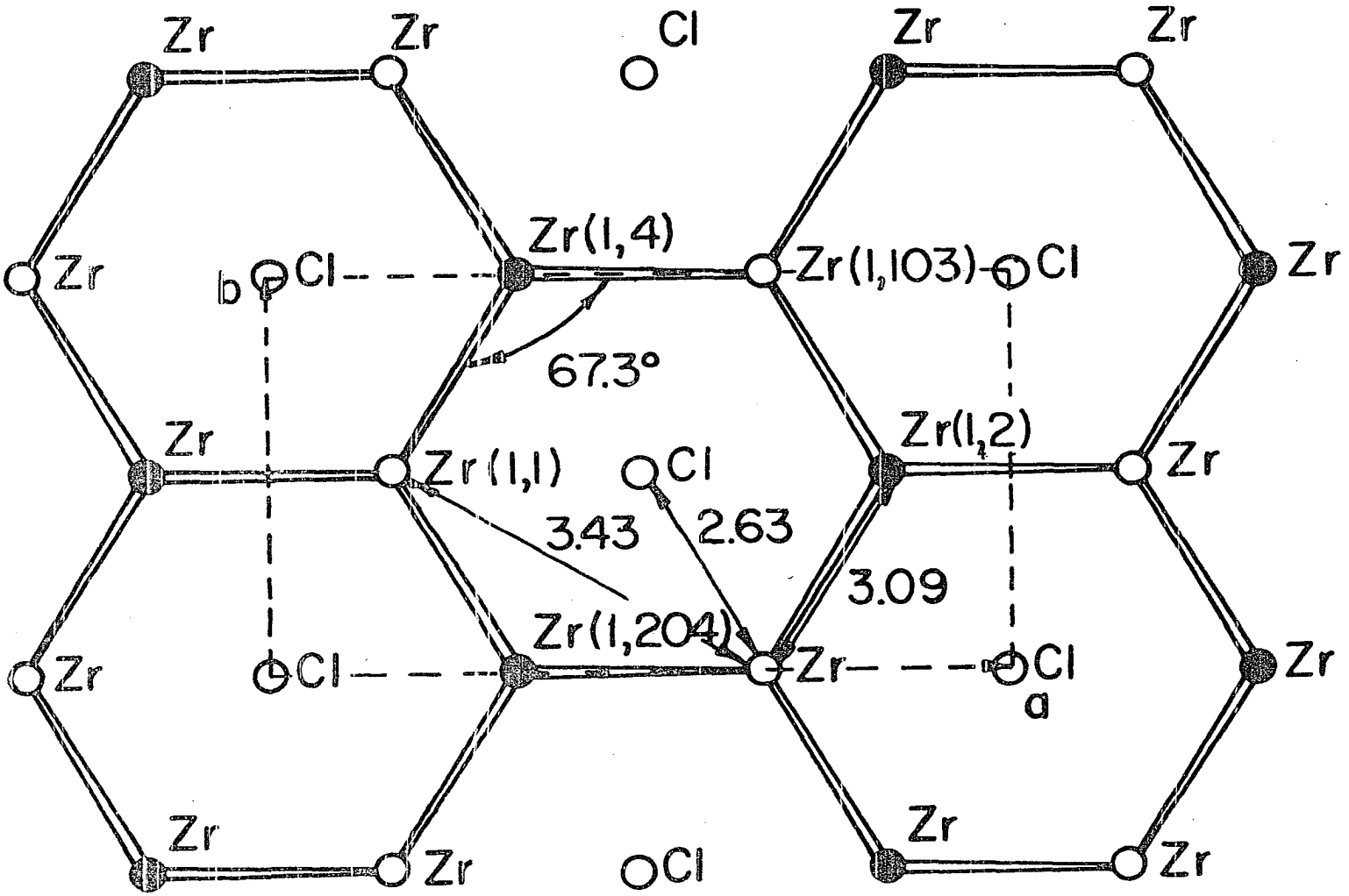


Figure 1. Projection of the structure of ZrCl on the (010) plane. Atoms at y=0.0 are represented by open circles while those at y=0.5 are shaded.

Figure 2. Projection of the structure of ZrCl on the (001) plane. Zirconium atoms at z=0.1341 are shaded while those at z=-0.1341 are represented by open circles. Chlorine atoms at z=±0.3296 are superimposed.


Zr
Zr
Cl
Zr
Zr
Zr
Zr(1,4)
Zr(1,103)
Cl
Zr
b
Cl
67.3°
Zr
Zr(1,2)
Zr
Zr(1,1)
Cl
3.43
2.63
3.09
Zr(1,204)
Zr
Cl
Zr
Cl
a
Zr
Zr
Zr
Cl
Zr
Zr

standard deviations were calculated using the variance-covariance matrix from the final least squares cycle. These are tabulated for the anisotropic refinement using new weights in Table 6.

Table 6. Interatomic distances and angles[a] from anisotropic refinement of ZrCl

Interatomic Distances[b]		Interatomic Angles[c]			
Intralayer		Atom 1	Atom 2 (Vertex)	Atom 3	Degrees
Zr-Zr	3.419(3) Å				
	3.428(3)	Zr(1,1)	Zr(1,4)	Zr(1,103)	67.30(15)
		Zr(1,204)	Zr(1,4)	Zr(1,2)	60.09(3)
Cl-Cl	3.419(3)	Zr(1,4)	Zr(1,2)	Zr(1,104)	120.18(7)
	3.428(3)	Zr(1,4)	Zr(1,2)	Zr(1,101)	124(4)
		Zr(1,4)	Zr(1,2)	Zr(1,103)	56(4)
Interlayer		Zr(1,101)	Zr(1,2)	Zr(1,103)	67.30(15)
Zr-Zr	3.09(14)	Zr(1,104)	Zr(1,2)	Zr(1,103)	90(5)
	3.09(14)	Zr(1,104)	Zr(1,2)	Zr(1,101)	56(4)
Cl-Cl	3.61(13)	Cl(2,2)	Zr(1,2)	Zr(1,104)	131(3)
	3.61(17)	Cl(2,2)	Zr(1,2)	Zr(1,101)	171(1)
		Cl(2,2)	Zr(1,2)	Zr(1,103)	105(1)
Zr-Cl	2.63(13)	Cl(2,2)	Zr(1,2)	Zr(1,4)	49(3)
	2.63(11)	Cl(2,2)	Zr(1,2)	Cl(2,104)	81.3(4)
		Zr(1,2)	Cl(2,2)	Zr(1,4)	81.3(4)

[a]Estimated standard deviations from ORFFE[65] are in parentheses.

[b]Differences of pairs of distances are statistically insignificant.

[c]Reference numbers correspond to those shown in Figures 1 and 2.

Discussion

The remarkable layer structure found for ZrCl consists of approximately cubic-close-packed layers of either metal or chlorine in the sequence Cl-Zr-Zr-Cl. Each zirconium has three metal neighbors in the next layer at 3.09 Å, six metal neighbors in the same layer at 3.43 Å and three chlorine neighbors in the opposite layer at 2.63 Å. In zirconium metal there are twelve equal metal-metal distances of 3.19 Å. In Zr-Cl intralayer interatomic distances for chlorine are also 3.43 Å. Interlayer chlorine-chlorine distances are 3.61 Å. The sum of the usual atomic radii[7] is 3.62 Å, but chlorine-chlorine approaches as close as 3.22 Å have been reported in $GdCl_3$ and Gd_2Cl_3.[11] The zirconium chlorine distances of 2.63 Å compare with three zirconium-chlorine distances 2.498, 2.655 and 2.307 Å found for $ZrCl_4$ [66] and a zirconium-chlorine distance of 2.44 Å in Rb_2ZrCl_6.[67]

The short zirconium-zirconium interlayer distance suggests a strong interaction between zirconium sheets. Another measure of bond formation suggested by Pauling[68] involves calculating the bond order n from the equation

$$D(n) = D(1) - 0.60 \log n$$

where D(1) is twice the single bond radius. Using a single bond radius of 1.454 Å for zirconium,[7] a bond order of 0.50 is obtained for the 3.09 Å interlayer distance while that for 3.43 Å intralayer distance is 0.13. For a chlorine

single bond radius of 0.99 Å[87] a bond order of 0.023 is obtained for the 3.43 Å chlorine-chlorine intralayer distance and 0.005 for the 3.62 Å interlayer distance. The bond order for the zirconium-chlorine distance is 0.48. The sum of bond orders to a particular atom is given by $\Sigma N_i n_i$ where N_i is the number of bonds and their bond order is n_i. Thus, the sum of all bond orders to zirconium is 3.7 and to chlorine is 1.4. Similar bond orders and distances were found for Hf and S in Hf_2S by Franzen and Graham.[69] In Hf_2S each S atom is surrounded by six Hf atoms in trigonal prismatic coordination. The coordination polyhedron around each Hf is a distorted octahedron of three S and three Hf atoms. Around each S atom there are six equivalent Hf-S distances of 2.63 Å with a bond order of 0.56 and six S-S distances of 3.37 Å with bond order 0.008. Around each Hf atom there are three Hf-Hf distances of 3.06 Å with bond order 0.50 and six Hf-Hf distances of 3.37 Å with bond order 0.15. The sum of the bond orders to Hf is 4.08. A sum of bond orders of 3.36 to sulfur indicates substantial participation of 3d orbitals in bonding. It has been pointed out that the similar metal-metal and nonmetal-nonmetal nonbonded distances found in both structures may be dictated by close packing of the larger anions and may thus not necessarily be indicative of bond order.[70]

During the course of this structural investigation a structure for ZrCl was published by Troyanov.[26] Troyanov and

T'sirelnikov[25] had previously reported the synthesis of ZrCl and a powder pattern which agreed with that of Struss and Corbett[13] except for the omission of a weak line with d=9.73 Å. Troyanov and T'sirelnikov indexed the powder pattern of ZrCl on the basis of a rhombohedral lattice having a=9.12 Å, α=21.62° and Z=2. Troyanov proceeded with a single crystal structural determination using film methods. The choice of the space group $R\bar{3}m$ based on the improper indexing of the powder pattern is more likely the reason for the poor refinement he obtained than errors made in measuring the intensities from the films as he suggested. In any case the R factor did not drop below 0.29 Surprisingly enough the basic ordering and packing of layers Cl-Zr-Zr-Cl was properly deduced. The interatomic distances found of 3.41 Å for either Zr-Zr or Cl-Cl within a layer are reasonable, but interlayer distances of 2.87 Å for Zr-Zr, 2.81 Å for Cl-Cl and 3.10 Å for Zr-Cl are certainly unreasonable.

The layer structure found for ZrCl accounts well for the physical properties of the compound. Zirconium monochloride occurs as shiny black hexagonal platelets which are graphitic in character. Dean[24] has suggested the use of "Zirklor" as a lubricant; however, the use of a reduced compound for such a purpose is questionable. The reason for the difficulty encountered in finding a single crystal of ZrCl suitable for a structure determination is also apparent now that the

structure is known. The broad faces of the hexagonal platelets are parallel to the (001) planes whereas the thin dimension is roughly coincident with the c-axis. While there are strong interactions between Zr-Zr planes and Zr-Cl planes, the Cl-Cl interplanar distance of 3.62 Å indicates only weak van der Waals contacts. Therein lies the basis for the graphitic character of the compound and its propensity toward polycrystallinity.

Recently, ZrBr has been prepared in this laboratory from the reaction of $ZrBr_4$ and Zr turnings in a tantalum container heated in the final stages to 800° for 12 days.[23] ZrBr occurs as dark shiny platelets much like those of ZrCl although some needles of ZrBr have been observed. The powder pattern of ZrBr has been indexed by comparison with the powder pattern and the structure determined for ZrCl. Reasonable agreement between calculated and observed powder patterns has been obtained for ZrBr in the case where the coordination polyhedron around zirconium is a trigonal antiprism as in ZrCl; but the Br-Zr-Zr-Br four-layer sheets adopt an alternate packing to that of the Cl-Zr-Zr-Cl four-layer sheets in ZrCl.[23]

Preliminary measurements have shown that ZrCl reacts reversibly with hydrogen even at room temperature to form a golden phase approximating $ZrClH_{0.5}$.[71] Perhaps hydrogen enters the lattice filling the interstices between planes of

chlorines. No structural work has as yet been done, however, to confirm the hydrogen sites.

THE CRYSTAL STRUCTURE OF (4,7,13,16,21,24-HEXAOXA-1,10-DIAZABICYCLO[8.8.8]HEXACOSANE) SODIUM HEPTANTIMONIDE(-3)

Structure Determination

Structure solution

The heavy atoms were located using the program package MULTAN[72] which is a system for solving crystal structures using direct methods. In this approach an attempt is made to determine the phases of the structure factors without first deriving a set of atomic positions. Any structure determination involves many more observations than there are parameters. Thus all the structure factors cannot be independent. In 1948 Harker and Kasper showed that inequality relationships existed between the structure factors and could occasionally lead to definite information about the phases of structure factors.[73] The Cauchy inequality

$$\left|\sum_{j=1}^{N} a_j b_j\right|^2 \leq \left(\sum_{j=1}^{N} |a_j|^2\right)\left(\sum_{j=1}^{N} |b_j|^2\right)$$

was applied to the unitary structure factor

$$U(hk\ell) = \sum_{j=1}^{n} n_j \exp\{2\pi i(hx_j + ky_j + \ell z_j)\}$$

by letting $a_j = \sqrt{n_j}$ and $b_j = \sqrt{n_j}\exp\{2\pi i(hx_j + ky_j + \ell z_j)\}$. By taking the symmetry elements of a given space group into account, relationships among the larger unitary structure

factors can be deduced. For a centric cell, the signs of a number of structure factors can be chosen arbitrarily because of the existence of a number of nonequivalent centers of symmetry which may be chosen as the origin. With three-dimensional data the selection and arbitrary assignment of signs to three strong reflections amounts to fixing the origin of the cell. Once progress in sign determination breaks down because of the inability to relate fresh signs to ones already determined, sign symbols are introduced. At the end of the sign-determining process the signs of the largest U's should have been determined in terms of a few sign symbols. If the number of symbols is n, there are 2^n possible sets of signs. The structure is sought by calculating Fourier syntheses for each of these sets and evaluating the respective electron density maps. In general, inequality relationships can be used only to solve simple structures. By definition the maximum value of the unitary structure factor U(hkℓ) is 1. For N equal atoms in a centrosymmetric unit cell the average value of $|U(hk\ell)|^2$ is 1/N. As the complexity of the structure increases, the fraction of the structure factors for which the inequality relationships are applicable declines.

In 1952 Sayre,[74] Cochran[75] and Zachariasen[76] independently showed that even when the |U| values were smaller than necessary to satisfy the inequality relationships, the relationship

$$S(h)S(h')S(h-h') = +1$$

was probably true. Thus the sign S of $U(h+h', k+k', \ell+\ell')$ can be deduced from the known signs of $U(hk\ell)$ and $U(h'k'\ell')$ by taking the product of the signs of the latter two unitary structure factors. MULTAN[60] uses this relationship to develop a set of signs for the structure factors after first assigning arbitrary signs to a specified number of large $|U|$'s and relating the larger $|U|$'s using inequalities. A probability of correctness is given to each of the 2^n subsets arising from the n symbols used to relate the reflections. In the final step a Fourier synthesis is carried out on the most probable subset. The resulting electron density map is scanned automatically and the positional coordinates of the peaks are listed. Distances and angles among peaks within 1.9Å of each other are printed out in order to facilitate assessment of the reasonableness of the model. Each time the Fourier step is repeated a Fourier synthesis is carried out on the next most probable subset.

In the present structure solution the seven strongest peaks output from the Fourier step of MULTAN[60] were located in a cluster having interpeak distances on the order of 2.8Å. Following the assignment of these seven peaks as antimony atoms, three cycles of least squares refinement using ORFLS[60] resulted in a residual of 0.398. An electron density map calculated using the program ALFF[59] contained, in addition to the seven antimony atoms, three peaks of appropriate height for sodium atoms with smaller peaks appropriate to the

constituent carbon nitrogen and oxygen atoms of 2,2,2-crypt clustered about them. The 26 independent nonhydrogen atoms of the first crypt were readily located from a difference electron density map[1] phased by the seven antimony and three sodium atoms. The complete sets of atoms making up the other two independent crypts proved to be much more difficult to locate. Three cycles of least squares refinement of the 10 heavy atoms and the 26 atoms of the first crypt resulted in an agreement factor of 0.233. From a difference map phased with these 36 atoms, plausible positions for the 26 atoms of the second crypt were assigned. This turned out to be painstaking work as the programs used do not accommodate such a large problem well. After a cycle of least squares refinement was carried out on these 62 atoms, an agreement factor of 0.231 was obtained. Finally a third difference map was computed in order to locate the 26 independent atom positions of the third crypt. Locating the atoms of the final crypt proved to be even more difficult than for the second one. Only two thirds of the atoms comprising the molecule emerged in the difference map output by ORFURER.[77] Nitrogen and

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[1]Light atoms were located from electron density maps calculated using the program ORFURER.[77] ORFURER is an adaptation of the Fourier step from MULTAN[70] by M. Gifkins which calculates and searches electron density maps from the structure factors output from ORFLS.[60]

oxygen atom positions were assigned after comparing the peaks in the difference map with those in the electron density map phased by only the heavy atoms and calculated using ALFF.[59] Carbon atom positions were less readily assigned as the peaks were broad and not very large, and in some cases there was evidence for disordering of the ethylene bridges.

Refinement

The initial stages of the refinement were carried out using the block-diagonal least squares program FBLS.[78] The calculation time for FBLS is about one fourth of that required for the full matrix least squares program ORFLS,[60] and the refined parameters obtained from FBLS normally agree with those from ORFLS within the respective estimated standard deviations. Pertinent to this refinement of 88 independent atoms is the fact that FBLS can accept 100 atoms while the maximum number of atoms that can be accommodated by ORFLS is 83. The scattering factors used in either program were those of Hanson et al.[61] Scattering factors for antimony and sodium were corrected for both the real and imaginary components of anomalous dispersion.[52] In either program the reflections were weighted by σ_F^{-2} to account for the reliability of each measurement. In the initial stages of refinement a reduced dataset which had not been corrected for absorption was used. Refinement of all 88 atoms with isotropic temperature factors using FBLS resulted in converged

positional and thermal parameters with an unweighted residual of 0.172. After converting the isotropic temperature factors of the heavy atoms to anisotropic temperature factors, three cycles of block-diagonal least squares using FBLS with the dataset that had been corrected for absorption resulted in well-behaved positional and thermal parameters with weighted and unweighted residuals 0.1304 and 0.1676, respectively. A refinement of the parameters from FBLS using ORFLS[60] was attempted by refining only a part of the structure at a time after removing 5 carbon atoms from the atom list. Two cycles were carried out on the heavy atoms followed by two cycles on the first crypt, then two cycles on the second crypt and finally two cycles on the third crypt. Such a procedure is quite laborious as each cycle requires about 40 minutes of computer time, and jobs of this size do not enjoy daily turnaround at the computer center. This stepwise refinement resulted in converged parameters with R = 0.113 and R_w = 0.139. Conversion of the isotropic temperature factors of the light atoms of the first crypt to anisotropic temperature factors followed by two cycles of refinement using ORFLS did not produce a change in the unweighted residual. Thus the use of anisotropic temperature factors for the light atoms is unwarranted according to the statistical test of Hamilton.[62]

Improving the refinement

Since it was considered imperative to use all 88 independent nonhydrogen atoms in the final refinement, a Fortran version of ORFLS was recompiled with atom arrays dimensioned at 120. After this had been done it was discovered that a 95 atom version of ORFLS was resident on disk under the program name ORFLSA.[79] Thereafter, the final refinement cycles were carried out using ORFLSA with all 88 independent nonhydrogen atoms in the atom list.

The dataset being used in this structure solution was collected just prior to that collected from the crystal of ZrCl so it is probable that the same instrumental problems affect both datasets. However, when the dataset was reweighted using OMEGA,[64] two cycles of full matrix least squares refinement using ORFLSA on the 10 heavy atoms with all 78 light atoms in the atom list resulted in converged parameters for the heavy atoms with a residual of 0.121 and a weighted residual of 0.161. Since this represents an increase of 0.02 in the weighted agreement factor, the absorption-corrected dataset with original weights was used in the final steps of refinement.

Estimated standard deviations for bond distances and angles calculated using the converged parameters from ORFLS contained significant contributions from errors in the lattice constants. Therefore the lattice constants were redetermined prior to the final refinement. The crystal from which the

X-ray reflection dataset had been collected was reoriented on the diffractometer and tuned values of the angles 2θ, Ω, χ and ϕ were measured on both sides of the instrumental zero for 24 strong reflections with $17.5<2\theta<25°$. A least squares fit to twice the average Ω angle using the program LCR2[51] resulted in converged cell constants of a=23.292(7), b=13.791(6), c=25.355(6)Å and β=108.56(2)°. These compare with a=23.263(43), b=13.776(4), c=25.344(73)Å and β=108.57(19) obtained previously. One cycle of least squares refinement using ORFLSA with the more precise lattice constants was carried out on the ten heavy atoms with anisotropic temperature factors and on the nonhydrogen atoms of the first crypt with isotropic temperature factors. A file of the final parameters and the variance-covariance matrix was created in order to calculate bond distances and angles and their standard deviations. Two cycles of refinement on the nonhydrogen atoms of the second crypt with isotropic thermal parameters using ORFLSA resulted in converged parameters, and a file containing the final parameters and the variance-covariance matrix was created. It was not considered worthwhile to spend more time refining the third crypt since earlier refinements of this crypt using ORFLS had not resulted in improved standard deviations for the atomic parameters over those obtained from FBLS. Finally two cycles of refinement on the heavy atoms using ORFLSA with the refined light atom parameters as described above in the atom list resulted in

converged positional parameters for the heavy atoms with a conventional R of 0.111 and a weighted R of 0.134. The ratio of the largest shift to standard deviation in coordinate in the last refinement cycle was 0.07. A final calculation of structure factors was carried out after the last cycle. A difference Fourier map showed no features other than two ripples of less than 3 and 5 $e^-/Å^3$, respectively, in the vicinity of Sb(5) and one ripple of less than 3 $e^-/Å^3$ near Sb(6). This is not unreasonable since scattering factors for Sb^0 were used. Otherwise, the map showed only a randomly fluctuating background of $\leq \pm 1$ $e^-/Å^3$. Since no discernable hydrogen peaks appeared in the difference map, these atoms were not included in the refinement. The calculated and observed structure factors for all 4662 observed reflections are tabulated in Table 7.[1] The final atomic and thermal parameters for the 88 independent nonhydrogen atoms are listed in Table 8.

Interatomic distances and angles

Interatomic distances and angles for the Sb_7^{3-} anion and the first two crypts were calculated using the function and error program ORFFE.[66] Standard deviations were estimated using the variance-covariance matrix from the appropriate

[1]Structure factors shown in Table 7 have been multiplied by 10.

Table 7. Observed and calculated structure factors for $(NaC^{+})_3Sb_7^{3-}$

L = 0

H	K	FC	FC
4	0	1537	1436
6	0	1349	1369
8	0	459	384
10	0	638	401
14	0	757	752
16	0	552	393
18	0	456	497
20	0	1053	1188
22	0	334	374
24	0	332	147
1	1	761	689
2	1	244	417
3	1	577	602
4	1	957	1295
5	1	2958	3033
6	1	3917	3476
7	1	687	816
8	1	839	664
9	1	2356	2260
10	1	1124	887
11	1	2001	1914
13	1	493	395
15	1	512	525
16	1	661	679
18	1	716	727
0	2	2353	2361
1	2	4563	4376
2	2	1472	1416
3	2	1515	1091
4	2	1542	1361
5	2	2990	2840
6	2	775	792
7	2	1415	1216
8	2	1223	1363
9	2	864	731
11	2	1707	1367
12	2	867	668
13	2	787	815
14	2	294	372
18	2	367	269
19	2	439	357
1	3	1397	1372
2	3	656	589
3	3	4082	4005
4	3	1475	1511
5	3	4596	4215
6	3	1519	1339
7	3	2396	2580
8	3	2048	1937
9	3	2471	2601
10	3	3673	3436
11	3	1838	1786
12	3	1748	1570
13	3	953	1016
14	3	725	804
15	3	483	619
16	3	775	772
1	4	1530	1054
2	4	1383	1221
4	4	375	362
5	4	507	561
6	4	1128	993
7	4	666	560
8	4	1120	952
9	4	1315	1424
10	4	515	483
11	4	1325	1131
12	4	1089	1005
13	4	949	816
15	4	1006	924
16	4	661	531
18	4	377	430
19	4	384	531
21	4	527	578
1	5	582	613
2	5	741	733
3	5	1808	1681
4	5	1617	1602
5	5	424	461
6	5	1191	1193
8	5	287	318
9	5	1569	1367
10	5	515	502
11	5	924	1090
13	5	527	468
14	5	443	286
15	5	625	567
16	5	415	358
0	6	478	647
1	6	2471	2328
2	6	2505	2584
3	6	2361	2226
4	6	4217	4094
5	6	1410	1464
6	6	3307	3052
8	6	754	723
9	6	349	220
12	6	414	353
14	6	1080	1045
15	6	555	409
16	6	655	760
20	6	427	426
1	7	368	518
2	7	1497	1491
3	7	1016	1002
4	7	1811	1749
5	7	1294	1206
6	7	373	228
7	7	543	563
8	7	654	663
11	7	726	617
12	7	754	645
13	7	531	493
14	7	728	671
15	7	684	709
16	8	959	1142
1	8	783	665
2	8	1072	1118
4	8	459	528
5	8	361	471
7	8	893	749
8	8	451	291
9	8	1089	1084
10	8	422	381
11	8	367	303
13	8	391	198
14	8	386	300
15	8	542	513
1	9	381	447
2	9	782	724
3	9	964	887
4	9	1764	1741
5	9	717	687
6	9	1546	1518
7	9	834	754
8	9	736	613
9	9	1076	914
10	9	363	355
11	9	402	368
14	9	597	631
16	9	480	517
1	10	550	559
2	10	355	143
3	10	418	359
5	10	1073	1018
6	10	412	333
7	10	911	899
9	10	609	684
11	10	357	265
12	10	545	497
17	10	352	212
5	11	703	694
7	11	609	667
0	12	1066	1059
1	12	1069	889
2	12	735	646
3	12	605	580
5	12	527	466
8	13	329	9
3	16	355	126

L = 1

H	K	FO	FC
1	0	2621	2188
5	0	3287	2496
7	0	1581	1307
9	0	1727	1945
11	0	1845	1741
13	0	520	596
15	0	977	928
17	0	1354	1461
19	0	574	533
0	1	3344	3197
1	1	4519	4096
2	1	1680	1666
3	1	3198	2931
4	1	670	789
5	1	970	929
6	1	343	207
7	1	1344	1717
8	1	1002	1164
9	1	980	1277
10	1	854	944
11	1	779	1008
12	1	1466	1439
13	1	410	462
18	1	722	673
19	1	467	558
20	1	507	593
0	2	2264	2037
1	2	568	663
2	2	2049	2210
3	2	1079	990
4	2	6372	6047
5	2	478	438
6	2	3866	3815
7	2	3158	3083
8	2	1762	1717
9	2	4079	3859
10	2	625	564
11	2	2702	2544
12	2	839	874
14	2	811	988
15	2	945	843
17	2	781	775
19	2	375	309
0	3	803	559
1	3	1715	1450
2	3	306	397
4	3	265	212
5	3	1478	1415
6	3	1110	972
10	3	1411	1238
11	3	432	365
12	3	1584	1550
14	3	370	256
15	3	600	703
19	3	510	455
20	3	527	469
0	4	979	827
2	4	3092	2895
3	4	2607	2286
4	4	1281	1162
5	4	2907	2816
6	4	1868	1881
7	4	800	976
8	4	3938	4047
9	4	1869	1757
10	4	3306	3116
11	4	995	1037
12	4	1110	1149
13	4	342	405
14	4	359	151
16	4	575	493
0	5	416	350
2	5	541	791
3	5	1351	1366
4	5	3095	3128
5	5	2319	2118
6	5	516	694
7	5	1239	1106
8	5	895	877
10	5	963	884
11	5	725	836
12	5	936	863
13	5	975	1079
14	5	1560	1600
15	5	462	516
16	5	1017	1032
20	5	364	365
21	5	503	461
0	6	492	511
1	6	398	463
2	6	713	722
3	6	747	680
4	6	700	718
5	6	518	542
6	6	414	552
8	6	536	525
12	6	1267	1360
13	6	1078	1120
17	6	357	445
0	7	1447	1539
1	7	1536	1617
3	7	2289	2383
4	7	346	237
5	7	1436	1381
8	7	719	714
9	7	498	441
11	7	726	688
13	7	1188	1126
15	7	888	964
16	7	372	389
0	8	342	200
1	8	941	982
2	8	611	671
3	8	2126	2041
4	8	323	367
5	8	1928	1973
7	8	1101	1053
8	8	653	630
10	8	570	673
14	8	256	379
6	9	1226	1393
9	9	1090	1073
11	9	373	282
13	9	360	359
2	10	654	789
3	10	506	459
4	10	1298	1432
5	10	826	860
6	10	1422	1410
7	10	560	565
8	10	813	880
13	10	440	539
0	11	1064	917
1	11	392	417
2	11	681	448
5	11	369	429
7	11	538	641
10	11	396	416
12	11	440	383
13	11	483	361
0	12	326	244
1	12	466	339
2	12	398	362
5	12	500	486
1	13	660	544
3	13	388	302
11	13	379	285
2	15	378	196

L = 2

H	K	FO	FC
0	0	6350	6599
2	0	7184	6698
4	0	1434	974
6	0	1190	1341
8	0	963	1094
10	0	2314	2369
12	0	841	1070
14	0	381	270
16	0	365	461
18	0	1394	1529
20	0	733	964
24	0	348	245
0	1	1659	1550
1	1	1682	1292
2	1	3156	3095
4	1	1322	916
5	1	1951	1710
6	1	976	1077
8	1	302	309
9	1	1718	1564
10	1	282	165
11	1	932	870
12	1	483	521
13	1	552	698
14	1	514	371
15	1	409	501
16	1	888	902
18	1	390	549
0	2	1369	1294
1	2	1642	1770
2	2	882	1010
3	2	3503	2567
4	2	2116	1708
5	2	2155	1532
6	2	827	831
7	2	330	314
8	2	619	766
9	2	1892	1589
11	2	1885	1768
12	2	576	531
19	2	467	415
20	2	345	248
23	2	429	320
0	3	2353	1904
1	3	1873	1813
2	3	692	604
3	3	4606	4316
4	3	2134	1816
5	3	4776	4743
6	3	519	473
7	3	4215	4049
8	3	3314	3100
9	3	2744	2486
10	3	2926	2891
11	3	1351	1603
13	3	748	763
14	3	870	925
16	3	539	578
25	3	299	63
0	4	539	643
2	4	1619	1648
3	4	2103	2120
4	4	604	744
5	4	1287	1477
6	4	1786	1668
8	4	1619	1609
9	4	834	673
11	4	907	644
12	4	473	456
13	4	1063	1069
15	4	984	850
16	4	498	532
19	4	559	679
21	4	376	394
0	5	417	292
1	5	829	723
3	5	282	520
4	5	534	530
6	5	288	141
7	5	1314	1230
9	5	1987	1910
11	5	904	965
12	5	371	414
13	5	486	514
14	5	903	1029
16	5	577	560
0	6	630	580
1	6	1711	1658
2	6	1822	1814
3	6	1302	1153
4	6	2569	2504
5	6	481	557
6	6	1212	1100
7	6	869	776
9	6	311	270

Table 7. (Continued)

10	6	930	875
11	6	553	506
12	6	1422	1382
13	6	736	691
14	6	1452	1666
15	6	511	583
16	6	511	578
20	6	353	413
0	7	418	622
1	7	592	525
3	7	1464	1419
5	7	1364	1435
7	7	377	397
9	7	669	792
11	7	492	438
12	7	691	796
13	7	408	430
14	7	362	447
15	7	472	682
21	7	319	85
0	8	370	317
1	8	628	363
2	8	636	550
3	8	315	305
5	8	459	495
7	8	655	650
12	8	539	575
13	8	389	405
15	8	480	496
1	9	1102	1241
2	9	1468	1543
3	9	908	887
4	9	1998	2151
5	9	717	751
6	9	1558	1563
7	9	1072	1082
8	9	316	281
9	9	859	901
11	9	496	363
1	10	576	505
2	10	336	336
3	10	333	405
5	10	426	457
6	10	448	484
10	10	486	433
12	10	558	570
14	10	314	97
1	11	328	160
9	11	324	493
0	12	842	741
1	12	514	356
2	12	448	234
11	12	413	449
0	13	483	441
2	13	646	543
4	13	460	358

L = 3

H	K	FO	FC
1	0	3220	2742
3	0	297	904
5	0	729	719
7	0	3142	2733
9	0	673	557
13	0	694	741
15	0	316	254
17	0	885	899
0	1	2957	2884
1	1	1933	1771
2	1	363	551
3	1	963	692
4	1	2038	1748
5	1	2147	1955
6	1	1199	1027
7	1	2071	1997
8	1	1392	1147
9	1	1539	1597
10	1	1488	1478
11	1	1336	1412
17	1	659	770
18	1	682	735
19	1	619	773
0	2	311	311
1	2	570	643
2	2	1731	1626
3	2	1371	1213
4	2	4230	3973
5	2	333	396
6	2	1750	1948
7	2	1091	1219
8	2	979	785
9	2	3037	3163
10	2	284	94
11	2	1568	1517
12	2	1131	1142
13	2	409	432
14	2	739	836
15	2	947	912
17	2	917	976
19	2	495	519
0	3	2478	1957
1	3	1028	1010
3	3	2810	2873
4	3	1444	1670
5	3	477	343
6	3	1533	1845
7	3	1142	1203
8	3	1215	1324
9	3	950	907
10	3	363	518
11	3	931	908
13	3	544	578
15	3	546	652
19	3	483	440
0	4	3129	2563
2	4	932	784
3	4	1728	1691
4	4	1369	1206
5	4	2337	2347
6	4	1037	1042
7	4	686	666
8	4	3103	2974
9	4	1349	1484
10	4	2370	2406
11	4	948	994
12	4	402	260
14	4	490	455
16	4	441	309
18	4	405	427
0	5	1151	1099
1	5	1696	1909
2	5	1298	1122
3	5	758	752
4	5	442	417
5	5	2299	2029
7	5	1531	1353
8	5	946	934
10	5	390	415
11	5	560	577
12	5	1069	1196
13	5	842	873
14	5	1704	1712
15	5	718	735
16	5	618	718
0	6	270	78
1	6	1343	1758
2	6	320	219
3	6	1347	1467
5	6	1813	1810
9	6	561	355
11	6	324	529
13	6	867	986
15	6	343	330
16	6	462	410
19	6	338	243
2	7	500	499
1	7	432	227
2	7	278	223
3	7	449	376
4	7	870	963
5	7	343	262
7	7	390	508
8	7	290	139
9	7	680	732
11	7	732	779
13	7	1367	1544
15	7	1012	1066
20	7	356	325
0	8	331	274
1	8	867	701
2	8	499	494
3	8	1518	1533
4	8	859	862
5	8	1074	998
7	8	628	480
8	8	1063	1067
10	8	893	765
11	8	448	493
12	8	343	335
1	9	493	359
2	9	475	526
3	9	619	545
10	9	464	521
12	9	376	368
0	10	467	478
1	10	321	269
2	10	471	536
3	10	560	632
4	10	970	1081
5	10	688	719
6	10	762	826
11	10	319	215
13	10	314	357
0	11	362	329
1	11	608	610
0	12	670	505
1	12	803	705
2	12	694	549
3	12	494	406
5	12	349	258
7	12	425	293
9	12	483	363
9	13	394	323

L = 4

H	K	FO	FC
0	0	293	261
2	0	2473	2860
4	0	4273	3728
6	0	2617	2320
8	0	3082	3055
10	0	1747	1754
16	0	456	607
18	0	1121	1344
20	0	516	618
0	1	2321	2456
1	1	827	832
2	1	4420	3971
3	1	2140	1910
4	1	3336	3310
5	1	611	666
6	1	487	491
7	1	1291	1229
8	1	622	529
9	1	940	935
10	1	285	485
11	1	735	731
12	1	542	655
13	1	1032	1051
14	1	290	211
15	1	416	570
16	1	800	610
0	2	1770	1540
1	2	579	507
2	2	964	727
3	2	1332	1058
4	2	1182	1138
5	2	634	754
7	2	429	851
9	2	371	96
10	2	559	398
11	2	476	376
13	2	676	739
15	2	441	584
17	2	675	574
0	3	1298	1180
1	3	3579	3307
2	3	1822	1885
3	3	1765	1600
4	3	3166	3150
5	3	1302	1436
7	3	1753	1817
8	3	3344	3238
9	3	1238	1258
10	3	1925	1901
11	3	962	859
12	3	613	557
14	3	601	588
15	3	471	414
16	3	517	464
17	3	518	567
18	3	421	376
0	4	360	686
1	4	1457	1278
2	4	646	658
3	4	2622	2529
4	4	952	766
5	4	1857	1965
6	4	1907	1901
7	4	1340	1303
8	4	1071	926
9	4	1088	1249
10	4	450	495
13	4	803	817
14	4	356	333
15	4	545	582
19	4	523	514
0	5	1356	1275
2	5	458	572
3	5	2094	2169
4	5	977	881
5	5	944	1051
7	5	1306	1223
8	5	322	266
9	5	1738	1569
12	5	395	398
14	5	399	432
16	5	304	263
0	6	3076	3349
1	6	881	1035
2	6	376	774
4	6	767	476
5	6	1000	1019
6	6	483	375
7	6	549	656
8	6	238	79
9	6	337	295
10	6	296	445
11	6	399	486
12	6	1442	1548
13	6	583	610
14	6	1414	1571
16	6	477	575
20	6	376	281
0	7	683	669
1	7	409	461
2	7	1100	1141
3	7	1141	1122
4	7	1051	1034
5	7	836	708
6	7	379	458
7	7	779	812
8	7	292	137
9	7	876	844
12	7	368	433
0	8	778	867
1	8	292	451
2	8	469	507
3	8	600	630
5	8	294	356
7	8	390	395
8	8	576	516
9	8	326	434
11	8	494	466
0	9	476	474
1	9	882	943
2	9	1084	1068
3	9	370	448
4	9	1551	1501
5	9	398	401
6	9	490	423
7	9	556	602
8	9	576	558
10	9	535	426
0	10	704	683
1	10	309	415
2	10	617	589
6	10	507	621
7	10	350	394
9	10	515	514
10	10	404	326
12	10	427	281
16	10	316	129
1	11	551	384
6	11	341	269
10	11	441	369
3	12	420	360
9	12	457	304
0	13	672	530
1	13	309	298
2	13	624	484
3	13	442	377
4	13	314	282

L = 5

H	K	FO	FC
1	0	1218	1541
3	0	806	511
5	0	546	452
7	0	3010	2839
9	0	916	868
11	0	1755	1872
13	0	1299	1425
15	0	519	463
17	0	367	287
0	1	636	533
1	1	4551	3941
2	1	4454	3918
3	1	2754	2641
4	1	3321	2811
5	1	3735	3292
6	1	300	110
7	1	2266	2151
8	1	1750	1561
9	1	1220	1281
10	1	735	597
12	1	825	830
13	1	775	777
14	1	754	792
17	1	813	880
18	1	418	500
19	1	505	582
0	2	489	470
1	2	687	773
2	2	2543	2313
3	2	3208	3242
4	2	2017	1750
5	2	1384	1410
6	2	807	834
8	2	1975	1888
9	2	675	649
10	2	653	723
12	2	1066	1047
13	2	874	822
15	2	769	814
17	2	455	544
0	3	1331	1123
1	3	886	778
2	3	303	119
3	3	1455	1313
5	3	656	607
6	3	2148	2279
7	3	975	862
8	3	1329	1494
9	3	376	349
10	3	1107	1146
12	3	997	1003
13	3	595	500
0	4	1766	1531
2	4	653	614
3	4	2079	1872

Table 7. (Continued)

4	4	1951	1849	8	9	841	809	15	2	401	438	1	7	757	740	0	1	1510	1234	8	5	899	931	10	11	350	206
7	4	1402	1400	10	9	744	826	17	2	355	355	2	7	640	504	1	1	3001	3152	9	5	497	496	12	11	394	408
8	4	450	340	13	9	399	331	19	2	345	229	3	7	877	911	2	1	2620	2431	10	5	375	212	10	12	333	255
9	4	1228	1376	0	10	747	723	1	3	720	704	4	7	813	669	3	1	1969	1813	11	5	419	519	1	13	583	482
12	4	469	558	2	10	403	359	2	3	3199	2964	5	7	373	338	4	1	736	793	13	5	565	670	3	13	326	222
14	4	705	709	3	10	664	740	3	3	938	763	7	7	1234	1277	5	1	1997	1948	15	5	359	359				
15	4	328	378	5	10	388	394	4	3	3478	3315	8	7	358	260	6	1	273	356	16	5	852	833		L =	8	
18	4	333	375	6	10	358	275	5	3	2257	2283	9	7	1048	1278	7	1	1320	1333	18	5	370	444	H	K	FO	FC
0	5	1366	1410	8	10	609	551	7	3	2303	2260	10	7	546	572	10	1	551	551	1	6	1337	1239	0	0	3145	3425
1	5	1275	1502	0	11	350	233	8	3	1226	1354	14	7	516	558	11	1	1025	912	2	6	460	330	2	0	2842	3129
2	5	528	675	1	11	469	451	9	3	1170	1178	16	7	578	562	12	1	1534	1580	3	6	1203	969	4	0	3819	4001
4	5	761	822	2	11	712	635	10	3	354	441	0	8	995	1036	13	1	1541	1526	6	6	379	383	6	0	2498	2362
5	5	1054	828	0	12	519	426	11	3	794	829	1	8	813	849	14	1	995	1088	7	6	969	1160	8	0	1350	1427
6	5	1011	1035	2	12	634	528	12	3	490	462	2	8	1185	1185	15	1	524	485	8	6	359	369	10	0	1746	1576
7	5	1468	1315	3	12	525	376	13	3	609	648	3	8	329	445	0	2	1178	1047	10	6	604	692	12	0	2826	2730
11	5	437	594	5	12	354	273	14	3	434	482	4	8	655	697	1	2	1920	1823	13	6	516	555	14	0	1286	1272
12	5	911	1006	7	12	481	408	16	3	436	457	7	8	313	325	2	2	2104	1892	15	6	700	673	0	1	965	984
13	5	758	1012	1	13	506	493	18	3	558	511	8	8	543	398	3	2	4175	3907	17	6	415	368	1	1	2506	2292
14	5	612	686	3	13	411	404	0	4	1443	1441	9	8	765	740	4	2	963	1017	0	7	359	387	2	1	427	98
15	5	618	601	3	14	362	238	1	4	837	756	10	8	579	530	5	2	2912	2847	1	7	2226	2197	3	1	2328	2082
18	5	410	422					2	4	1040	1040	11	8	416	501	6	2	2760	2659	2	7	496	495	4	1	1067	1070
1	6	1504	1314		L =	6		3	4	407	423	17	8	387	403	8	2	1912	2103	3	7	1410	1674	5	1	375	372
3	6	2660	2307	H	K	FO	FC	4	4	1396	1287	2	9	704	749	10	2	339	348	4	7	352	350	6	1	2335	2322
4	6	297	280	0	0	5530	5284	5	4	1208	1307	3	9	397	412	11	2	470	518	5	7	991	998	7	1	1623	1605
5	6	973	1187	2	0	6969	5807	6	4	1414	1337	6	9	824	791	13	2	535	669	7	7	458	436	8	1	520	686
7	6	288	364	4	0	5455	5126	7	4	1749	1714	7	9	391	265	15	2	459	491	9	7	723	646	9	1	489	619
8	6	334	364	6	0	3849	3810	8	4	565	483	8	9	1254	1353	18	2	367	339	12	7	347	290	10	1	511	406
14	6	513	588	8	0	3464	3188	9	4	1553	1459	9	9	489	505	0	3	490	324	15	7	444	325	11	1	825	794
16	6	375	375	10	0	495	454	10	4	351	287	10	9	573	672	1	3	1493	1348	17	7	311	344	12	1	685	584
17	6	426	373	12	0	1913	1817	11	4	757	717	11	9	432	447	3	3	885	900	1	8	273	336	16	1	399	375
0	7	554	594	14	0	940	814	12	4	455	383	0	10	983	893	4	3	1643	1679	2	8	642	634	17	1	363	420
1	7	1733	1824	18	0	622	711	13	4	329	402	2	10	924	821	5	3	584	584	4	8	368	393	18	1	396	326
2	7	504	614	0	1	1929	1869	16	4	348	427	6	10	409	451	6	3	1556	1614	5	8	683	692	0	2	939	743
3	7	1400	1552	1	1	652	703	17	4	527	616	9	10	503	537	7	3	355	252	6	8	856	761	1	2	867	927
5	7	1051	1329	2	1	1898	2069	0	5	855	804	11	10	724	652	8	3	980	1087	7	8	1225	1079	2	2	841	781
6	7	314	368	3	1	1480	1399	1	5	1176	1038	13	10	446	521	10	3	957	916	8	8	568	639	3	2	1223	1176
8	7	435	385	4	1	2344	2292	2	5	1408	1318	1	11	332	424	11	3	637	580	9	8	1288	1337	4	2	1356	1426
9	7	307	54	6	1	3663	3440	3	5	1798	1697	5	11	372	454	12	3	682	633	11	8	636	646	6	2	681	763
11	7	369	538	7	1	1946	1887	4	5	714	755	7	11	597	581	13	3	531	647	15	8	354	186	7	2	312	217
12	7	339	219	8	1	1803	1815	5	5	1671	1489	1	12	761	674	14	3	325	446	0	9	847	879	9	2	529	586
13	7	781	930	9	1	1172	1054	6	5	1087	922	2	12	332	344	17	3	444	559	1	9	757	767	11	2	1984	1960
15	7	320	447	10	1	289	322	7	5	398	361	3	12	605	508	0	4	848	748	2	9	437	525	13	2	1343	1287
0	8	558	576	11	1	777	754	10	5	489	510	4	12	488	314	1	4	1158	1227	3	9	434	373	0	3	1073	1133
1	8	836	825	12	1	705	571	11	5	399	410	5	12	350	210	2	4	3424	3376	8	9	821	859	1	3	1623	1486
2	8	933	852	13	1	944	958	12	5	477	601	1	13	357	284	3	4	1138	977	10	9	625	704	2	3	3903	3850
3	8	515	594	17	1	408	438	15	5	368	409	3	13	384	338	4	4	2989	2734	11	9	347	357	3	3	2089	2259
4	8	1030	1021	0	2	923	905	16	5	350	266					5	4	638	663	12	9	361	406	4	3	1981	1991
6	8	397	377	1	2	4008	3275	0	6	3319	3401		L =	7		6	4	928	929	3	10	444	425	5	3	3341	3253
7	8	692	602	3	2	748	582	1	6	1109	1323	H	K	FO	FC	7	4	1713	1681	4	10	541	577	6	3	385	450
8	8	1062	1071	4	2	399	384	2	6	2237	2193	1	0	1162	800	8	4	604	591	6	10	966	1014	7	3	3141	2865
9	8	953	1085	5	2	277	674	3	6	1747	1753	3	0	1209	1291	9	4	1600	966	7	10	332	378	8	3	353	394
10	8	479	568	6	2	296	207	4	6	1193	1472	5	0	1801	1905	10	4	1266	1284	8	10	774	852	9	3	2006	1954
11	8	566	574	7	2	318	86	5	6	1164	1262	7	0	1822	1733	12	4	929	973	10	10	476	446	10	3	309	536
0	9	526	605	8	2	354	487	7	6	475	596	9	0	352	436	0	5	2325	2214	0	11	643	561	11	3	820	883
1	9	782	755	9	2	669	671	8	6	385	214	11	0	1631	1455	1	5	2198	2326	1	11	392	291	12	3	538	568
2	9	432	368	10	2	591	592	12	6	652	842	13	0	961	966	2	5	663	740	2	11	602	530	13	3	649	736
3	9	610	537	11	2	1548	1640	14	6	627	673	15	0	648	631	4	5	855	1005	3	11	322	215	0	4	1760	1840
5	9	372	286	12	2	314	194	18	6	407	462	17	0	686	667	5	5	484	351	4	11	346	228	1	4	639	762
7	9	526	586	13	2	1501	1508	0	7	473	325	19	0	491	563	7	5	966	720	5	11	320	378	2	4	374	277

Table 7. (Continued)

3	4	445	290	5	9	451	396	5	4	2065	1894		L = 10			17	4	338	353	3	1	1334	1032	1	7	781	827
4	4	319	239	6	9	1244	1265	1	4	568	526	H	K	FO	FC	1	5	660	620	4	1	590	537	3	7	481	561
5	4	359	461	7	9	726	654	2	4	2781	2613	0	0	815	623	4	5	805	663	5	1	474	263	5	7	754	813
6	4	1010	886	8	9	1153	1236	3	4	1162	1214	2	0	1669	1293	5	5	1062	1091	6	1	274	93	7	7	696	872
7	4	1111	1122	9	9	567	611	4	4	1655	1495	4	0	309	286	6	5	937	1176	9	1	745	632	10	7	325	379
9	4	795	821	10	9	507	578	5	4	632	679	6	0	614	835	7	5	341	368	10	1	1369	1424	0	8	759	839
11	4	300	189	11	9	335	355	6	4	592	448	8	0	799	465	8	5	501	499	11	1	1092	1034	1	8	513	419
14	4	454	524	0	10	654	789	7	4	809	860	10	0	2013	1964	9	5	496	573	12	1	577	760	2	8	399	460
16	4	455	532	2	10	476	737	8	4	504	524	12	0	2319	2257	0	6	268	173	16	1	313	340	3	8	685	779
17	4	410	523	11	10	353	390	10	4	1126	1048	14	0	981	889	1	6	388	361	0	2	999	855	4	8	478	538
0	5	311	391	13	10	392	390	12	4	493	509	0	1	1412	1414	4	6	495	544	1	2	1734	1655	6	8	486	505
1	5	1056	725	5	11	354	497	15	4	331	393	1	1	3072	2991	5	6	321	405	2	2	1140	1125	0	9	554	535
2	5	471	369	7	11	371	421	17	4	493	472	2	1	1021	1185	6	6	1211	1236	3	2	885	774	4	9	453	438
3	5	1329	1125	0	12	340	304	0	5	323	114	3	1	1136	1104	7	6	439	441	4	2	910	870	6	9	435	416
4	5	387	247	1	12	487	486	1	5	789	844	4	1	744	682	8	6	702	940	5	2	416	368	8	9	423	399
7	5	300	52	2	12	419	360	6	5	897	932	5	1	741	571	11	6	318	349	6	2	862	969	2	11	326	195
9	5	575	623	6	12	307	178	7	5	536	493	6	1	341	381	16	6	434	594	7	2	692	732	5	11	384	334
10	5	308	169					8	5	719	764	7	1	767	714	0	7	526	641	8	2	461	460	1	12	498	410
11	5	598	664		L =	9		11	5	427	470	8	1	313	315	1	7	545	606	9	2	655	784	6	12	325	305
15	5	321	291	H	K	FO	FC	13	5	459	588	10	1	684	577	2	7	859	881	11	2	492	476	1	13	370	274
16	5	499	492	1	0	2835	2429	14	5	463	534	0	2	1785	1491	4	7	1284	1342	0	3	885	1002				
18	5	374	275	3	0	1945	2025	16	5	814	826	2	2	1124	968	5	7	777	761	1	3	900	865		L =	12	
0	6	2133	2069	5	0	1227	1253	0	6	593	512	3	2	900	965	6	7	793	1020	2	3	624	712	H	K	FO	FC
1	6	908	891	7	0	1118	1097	3	6	1036	992	4	2	523	693	7	7	773	746	4	3	359	219	0	0	2881	2574
2	6	1792	1696	9	0	1189	1089	5	6	1473	1516	5	2	287	293	9	7	354	368	5	3	263	270	2	0	3683	3314
3	6	1130	1114	11	0	779	737	7	6	1030	1248	6	2	473	470	10	7	516	459	6	3	410	412	4	0	1095	847
4	6	1388	1042	13	0	456	416	8	6	644	592	7	2	741	650	12	7	276	144	8	3	807	794	6	0	253	139
5	6	902	796	15	0	421	280	10	6	374	367	8	2	596	669	14	7	311	185	9	3	689	737	8	0	474	473
6	6	350	423	17	0	417	438	11	6	386	208	9	2	481	453	1	8	318	455	10	3	730	613	10	0	1158	1246
8	6	742	820	0	1	1348	1301	15	6	392	374	10	2	597	640	7	8	480	489	11	3	674	650	12	0	649	747
9	6	411	341	1	1	1027	1243	0	7	581	588	11	2	824	945	11	8	343	352	12	3	707	570	0	1	846	1045
10	6	360	175	3	1	256	372	1	7	701	651	12	2	357	251	1	9	461	446	14	3	374	387	1	1	1773	1743
12	6	346	424	5	1	694	866	2	7	550	591	13	2	365	494	2	9	307	256	15	3	512	580	2	1	855	920
13	6	549	557	6	1	517	397	3	7	836	792	14	2	402	279	4	9	375	397	0	4	2020	1939	3	1	634	684
15	6	533	481	7	1	1554	1291	7	7	843	869	16	2	321	367	5	9	365	332	1	4	876	885	5	1	1075	1032
16	6	705	625	8	1	310	349	8	7	322	228	17	2	337	229	6	9	665	609	2	4	658	718	6	1	561	664
0	7	738	808	9	1	692	568	9	7	620	664	0	3	2462	2245	8	9	416	390	3	4	657	559	7	1	337	181
1	7	744	766	10	1	1455	1608	15	7	451	555	1	3	1355	1253	9	9	340	423	4	4	686	642	8	1	1257	1362
3	7	536	547	11	1	1590	1585	0	8	498	419	2	3	2743	2456	12	9	294	27	6	4	861	910	10	1	496	572
4	7	583	772	12	1	1560	1651	1	8	464	524	3	3	2037	1767	0	10	340	378	11	4	299	9	11	1	360	302
5	7	685	700	13	1	1204	1132	2	8	396	416	4	3	465	385	3	10	480	472	17	4	317	320	13	1	291	70
6	7	536	608	15	1	331	369	5	8	398	391	5	3	1202	1139	4	10	456	421	0	5	1740	1741	0	2	1273	1205
7	7	1261	1428	0	2	1047	991	6	8	684	732	6	3	739	745	5	10	626	547	1	5	255	171	1	2	1012	1120
9	7	815	924	1	2	2578	2386	7	8	948	975	7	3	1157	1123	6	10	277	188	2	5	710	852	2	2	1171	1029
14	7	495	507	2	2	860	754	9	8	782	779	8	3	443	373	7	10	355	356	4	5	635	806	3	2	605	645
16	7	397	379	3	2	3111	2918	10	8	482	393	9	3	731	728	1	11	354	181	5	5	514	553	4	2	454	347
0	8	602	532	4	2	965	924	0	9	942	1015	11	3	548	533	0	13	484	443	6	5	1053	1172	5	2	636	572
1	8	553	608	5	2	710	767	1	9	368	382	16	3	467	383	2	13	439	305	10	5	567	425	7	2	575	388
2	8	934	908	6	2	2652	2562	4	10	455	528	17	3	328	276					11	5	482	594	8	2	755	851
4	8	374	366	7	2	405	352	6	10	727	740	0	4	1521	1505		L =	11		13	5	340	326	10	2	893	906
7	8	607	642	8	2	1202	1303	7	10	330	305	1	4	578	680	H	K	FC	FC	17	5	329	288	13	2	575	538
8	8	503	529	10	2	326	328	8	10	719	636	2	4	856	943	1	0	4402	4190	0	6	699	712	15	2	498	531
9	8	393	538	0	3	362	494	5	11	369	464	3	4	562	691	3	0	670	671	1	6	475	339	16	2	384	265
10	8	423	441	1	3	1447	1344	7	11	386	417	4	4	382	255	7	0	578	598	2	6	844	909	0	3	1986	1902
12	8	432	458	3	3	324	321	1	12	535	444	6	4	432	321	9	0	809	831	3	6	2111	1911	1	3	716	756
0	9	337	116	4	3	974	959	3	12	461	324	9	4	557	367	11	0	696	721	4	6	701	755	2	3	1546	1422
1	9	437	465	11	3	911	817	8	12	320	206	11	4	1021	956	0	1	1217	1296	5	6	2274	2305	3	3	419	314
3	9	673	720	15	3	484	593					14	4	430	590	1	1	2886	2538	7	6	976	933	4	3	661	493
4	9	386	192	17	3	400	444					15	4	456	475	2	1	754	769	8	6	389	426	5	3	569	571

Table 7. (Continued)

6	3	1153	1244
7	3	1209	1269
8	3	538	615
9	3	642	674
10	3	474	456
0	4	1083	1138
1	4	415	489
2	4	295	586
3	4	838	870
4	4	667	663
5	4	789	855
6	4	416	501
7	4	500	357
9	4	1256	1218
10	4	318	293
11	4	1278	1224
13	4	450	421
15	4	305	289
0	5	514	421
2	5	904	827
3	5	1017	1053
4	5	1247	1234
5	5	1437	1328
6	5	1230	1179
7	5	551	518
8	5	612	617
13	5	287	116
0	6	967	911
1	6	474	558
2	6	1028	981
3	6	782	704
4	6	1354	1467
5	6	760	628
6	6	1395	1324
7	6	389	429
10	6	405	461
1	7	733	806
2	7	977	1092
4	7	1338	1330
5	7	434	379
6	7	705	614
11	7	371	318
0	8	369	431
9	8	416	390
11	8	338	376
0	9	354	312
1	9	269	143
2	9	342	373
10	9	329	339
3	10	445	426
5	10	513	569
7	10	526	529
1	11	328	323
1	12	567	357
2	12	302	204
3	12	359	301

L = 13

H	K	FO	FC
1	0	1645	1705
3	0	355	230
5	0	331	128
7	0	300	412
9	0	363	497
0	1	775	915
1	1	3233	2947
2	1	1280	1213
3	1	1777	1512
6	1	532	526
9	1	382	362
10	1	345	548
14	1	524	537
16	1	356	372
0	2	1357	1280
3	2	603	715
5	2	994	1187
7	2	1146	1286
8	2	364	380
9	2	762	891
11	2	572	695
1	3	967	1028
3	3	299	287
4	3	594	497
5	3	280	432
6	3	787	747
7	3	304	350
8	3	782	633
9	3	853	909
10	3	980	968
11	3	308	433
12	3	598	633
0	4	862	835
1	4	459	512
2	4	549	487
4	4	1829	1691
5	4	327	374
6	4	1322	1469
7	4	469	509
8	4	766	816
10	4	358	454
0	5	1436	1368
2	5	1274	1300
3	5	765	717
4	5	1264	1205
5	5	820	664
6	5	668	831
7	5	436	488
9	5	386	413
10	5	778	816
11	5	470	607
12	5	360	248
0	6	555	500
2	6	998	866
3	6	1260	1234
4	6	649	599
5	6	1405	1320
7	6	394	464
13	6	337	327
1	7	1979	933
3	7	941	847
5	7	913	976
7	7	651	496
10	7	484	507
11	7	329	314
0	8	644	723
1	8	463	456
2	8	307	293
3	8	759	660
5	8	380	308
7	8	338	246
11	8	313	239
2	9	467	419
4	9	630	566
6	9	439	395
8	9	434	429
6	10	291	241

L = 14

H	K	FO	FC
0	0	3374	3157
2	0	1587	1590
8	0	524	372
14	0	677	729
16	0	463	396
0	1	1331	1344
1	1	475	541
3	1	533	476
5	1	643	709
6	1	504	537
8	1	803	862
10	1	529	583
0	2	572	562
1	2	451	493
2	2	417	370
4	2	391	300
5	2	671	568
7	2	312	234
8	2	463	553
9	2	306	313
10	2	550	607
11	2	384	276
13	2	613	633
15	2	506	473
0	3	804	847
1	3	531	495
3	3	910	940
4	3	868	865
5	3	1535	1453
6	3	473	470
7	3	1204	1209
8	3	506	604
9	3	1013	1050
10	3	431	587
11	3	677	526
2	4	398	369
3	4	592	568
5	4	520	638
7	4	410	428
9	4	1014	1082
10	4	426	275
11	4	785	785
13	4	313	212
14	4	315	136
0	5	487	384
2	5	551	564
3	5	1322	1211
4	5	1225	1309
5	5	885	834
6	5	854	932
0	6	1181	1006
1	6	614	453
2	6	1030	850
3	6	413	303
4	6	1305	1205
5	6	330	301
6	6	844	702
10	6	561	640
1	7	919	845
2	7	623	671
4	7	911	863
7	8	407	411
9	8	499	531
11	8	332	171
0	9	486	433
2	9	321	288
5	10	461	434
0	12	385	235
1	12	474	401

L = 15

H	K	FC	FC
1	0	592	553
3	0	1730	1509
5	0	1048	1015
0	1	290	337
1	1	1123	1078
3	1	319	251
4	1	780	672
12	1	482	488
14	1	587	551
0	2	808	774
3	2	454	390
5	2	998	1012
6	2	396	466
7	2	893	1072
8	2	606	458
9	2	614	713
11	2	309	328
15	2	343	241
0	3	1005	829
2	3	374	205
3	3	652	717
5	3	591	502
7	3	639	631
8	3	466	438
9	3	907	912
10	3	422	394
0	4	336	245
1	4	325	372
2	4	1058	949
4	4	1403	1358
6	4	1126	1035
8	4	703	669
10	4	394	339
2	5	621	564
3	5	626	494
4	5	972	906
5	5	748	633
7	5	329	304
8	5	640	682
10	5	738	756
12	5	336	244
1	6	452	327
3	6	424	456
11	6	328	311
1	7	277	55
3	7	551	404
5	7	452	249
8	7	374	309
11	7	325	265
0	8	512	567
1	8	414	462
3	8	394	346
1	9	374	315
2	9	531	466
4	9	336	299
6	9	317	323
0	10	362	396
0	12	332	141

L = 16

H	K	FC	FC
0	0	994	980
4	0	1087	1164
6	0	448	501
8	0	317	249
12	0	392	474
14	0	546	570
0	1	297	337
1	1	475	457
2	1	641	658
3	1	997	1019
4	1	498	437
5	1	420	405
1	2	645	575
2	2	427	407
3	2	879	804
5	2	690	773
10	2	291	184
11	2	304	171
0	3	367	341
1	3	616	510
2	3	666	481
3	3	687	564
4	3	581	501
5	3	672	714
6	3	367	279
7	3	437	460
9	3	384	280
14	3	369	340
0	4	372	379
5	4	401	296
6	4	472	601
8	4	753	674
9	4	452	645
1	5	292	254
3	5	349	393
8	5	290	51
10	5	309	286
4	6	581	443
6	6	318	159
8	6	449	518
10	6	423	480
0	7	327	280
1	7	631	685
0	8	447	501
2	8	585	514

L = 17

H	K	FC	FC
1	0	1326	1197
3	0	1936	1804
5	0	1297	1183
7	0	594	598
0	1	485	302
1	1	563	701
2	1	903	947
3	1	867	966
4	1	1056	998
5	1	394	482
0	2	323	246
1	2	306	75
2	2	327	453
4	2	404	357
12	2	338	232
0	3	893	768
2	3	319	369
3	3	530	426
6	3	342	263
7	3	449	545
9	3	365	520
11	3	302	83
0	4	286	181
2	4	475	416
4	4	366	382
0	5	792	759
3	5	450	399
5	5	479	470
8	5	656	661
10	5	335	301
1	6	595	408
3	6	355	34
5	6	419	321
6	6	470	400
7	6	512	464
1	7	530	671
1	9	376	410
0	10	326	199

L = 18

H	K	FO	FC
0	0	876	841
2	0	1320	1287
4	0	1140	1164
6	0	482	410
8	0	379	280
1	1	465	531
2	1	976	919
3	1	772	822
4	1	911	899
6	1	426	433
8	1	346	333
9	1	354	151
1	2	989	949
3	2	1011	959
9	2	366	348
0	3	330	142
1	3	521	426
1	4	381	218
6	4	652	731
8	4	543	568
5	5	463	425
7	5	322	295
10	5	336	244
0	6	622	640
1	6	453	422
0	7	329	133
4	7	454	304
6	7	410	393
0	8	575	684
2	8	464	495

L = 19

H	K	FO	FC
1	0	955	908
3	0	1016	1053
5	0	826	855
7	0	353	528
0	1	785	749
1	1	835	826
2	1	1032	997
3	1	1116	1073
4	1	816	755
5	1	467	439
2	2	362	434
3	2	434	396
5	2	366	488
6	2	382	451
7	2	492	364
0	3	430	442
1	3	344	323

Table 7. (Continued)

3	3	446	372
6	3	434	424
7	3	522	565
8	3	492	342
10	3	348	249
6	4	448	427
8	4	434	400
0	5	649	551
2	5	409	335
4	5	285	240
6	5	398	480
4	6	455	359
6	6	381	335
7	6	388	260
8	6	318	222
1	7	709	631
3	7	447	439
4	7	298	103
5	7	357	327
5	8	377	275
1	9	442	324
3	9	331	242

L = 20

H	K	FO	FC
0	0	979	1059
2	0	1223	1181
4	0	590	538
0	1	588	501
2	1	954	858
4	1	553	560
6	1	563	524
0	2	435	275
1	2	687	753
2	2	373	349
3	2	341	356
7	2	502	461
4	3	379	273
7	3	454	453
9	3	420	312
4	4	306	316
6	4	460	455
2	5	279	86
0	6	625	508
1	6	408	336
3	6	353	261
4	6	335	158
4	7	343	209
0	8	387	358
2	8	326	325

L = 21

H	K	FO	FC
3	0	413	339
9	0	327	111
0	1	497	521
2	1	571	516
3	1	318	293
6	1	425	277
3	2	437	379
1	3	461	489
7	3	339	343
4	4	314	299

L = 22

H	K	FO	FC
0	0	310	274
2	0	566	432
6	0	365	399
0	2	388	457
1	4	493	470
2	4	276	49
1	6	358	103
3	7	334	214
1	7	324	187

L = 23

H	K	FO	FC
0	3	472	365
1	3	452	513
2	3	381	366
0	5	379	338
1	6	368	290

L = 24

H	K	FO	FC
2	0	311	249
4	0	465	399
0	2	546	477
2	2	326	261
4	4	504	486

L = 25

H	K	FO	FC
1	1	317	281

L = -1

H	K	FO	FC
1	0	167	593
3	0	2840	2790
5	0	637	497
7	0	874	902
9	0	845	1037
11	0	3105	3372
13	0	1074	1327
15	0	577	765
17	0	614	514
0	1	3170	3197
1	1	5861	5550
2	1	4364	3906
3	1	5626	5410
4	1	307	225
6	1	2180	1982
7	1	377	676
8	1	526	450
9	1	445	375
10	1	485	561
12	1	585	484
13	1	1508	1535
15	1	1376	1431
16	1	640	615
17	1	740	846
20	1	560	672
22	1	410	445
0	2	1881	2037
1	2	353	552
2	2	1258	1066
3	2	620	765
4	2	551	721
5	2	556	494
6	2	3677	3055
7	2	1564	1413
8	2	976	902
9	2	2934	2613
10	2	1761	1675
11	2	2125	2160
12	2	964	808
13	2	575	415
14	2	484	628
15	2	504	658
16	2	754	700
17	2	421	455
21	2	378	342
23	2	357	210
0	3	704	559
1	3	1934	1897
2	3	2960	2424
3	3	2061	2049
4	3	2565	2082
5	3	1517	1324
6	3	1738	1365
7	3	1048	874
8	3	1529	1469
9	3	1485	1334
10	3	2437	2242
12	3	1490	1402
13	3	623	755
14	3	431	323
17	3	573	514
21	3	370	418
0	4	820	827
1	4	1100	1000
3	4	1332	1338
5	4	1650	1562
6	4	503	348
7	4	602	563
8	4	1397	1565
9	4	900	1038
10	4	1847	1648
11	4	1241	1073
12	4	1251	1270
13	4	528	482
14	4	445	282
15	4	640	678
17	4	462	440
18	4	435	488
0	5	444	350
1	5	1215	1257
2	5	1460	1403
3	5	375	270
4	5	2980	2952
5	5	2082	1815
6	5	1816	1822
7	5	2387	2347
9	5	633	538
11	5	324	331
13	5	334	307
16	5	463	504
19	5	561	531
21	5	477	497
0	6	526	512
1	6	2008	2351
2	6	1023	1008
3	6	1312	1353
4	6	344	414
5	6	389	330
7	6	923	824
8	6	863	708
9	6	669	644
10	6	663	620
11	6	425	198
12	6	714	666
13	6	861	809
14	6	615	618
15	6	1131	1077
17	6	372	333
0	7	1428	1540
1	7	717	708
2	7	967	828
3	7	2021	1925
5	7	2835	2628
7	7	1461	1460
9	7	365	433
10	7	498	404
11	7	394	339
14	7	434	400
16	7	521	566
18	7	375	300
19	7	373	395
0	8	276	200
1	8	581	625
2	8	477	499
3	8	1604	1570
5	8	1301	1256
6	8	962	892
7	8	753	714
8	8	432	240
10	8	647	621
11	8	425	286
12	8	450	394
14	8	471	462
17	8	620	624
0	9	292	194
1	9	541	586
2	9	466	239
4	9	1147	896
5	9	805	647
6	9	1540	1440
7	9	351	149
8	9	1605	1474
10	9	842	701
13	9	419	490
17	9	378	377
2	10	338	251
4	10	485	268
5	10	530	486
6	10	1008	804
7	10	524	517
8	10	617	535
11	10	440	243
0	11	978	917
1	11	558	547
2	11	735	543
7	11	614	619
13	11	413	327
1	13	845	719
3	13	456	312

L = -2

H	K	FC	FC
0	0	6436	6599
2	0	6312	5926
4	0	2015	1670
6	0	1065	857
10	0	3505	3216
12	0	2105	2218
14	0	1685	1740
16	0	1772	1913
18	0	547	446
20	0	556	525
22	0	510	529
0	1	1494	1550
1	1	205	203
2	1	1816	1562
3	1	2638	2508
5	1	1940	1837
6	1	3245	2276
7	1	1602	1484
8	1	2015	1537
9	1	1247	1269
10	1	385	430
11	1	3171	2930
12	1	713	556
13	1	632	568
15	1	408	502
17	1	334	268
22	1	390	363
0	2	1271	1294
1	2	2023	2079
2	2	425	987
3	2	1531	1369
4	2	1532	1159
5	2	3491	3535
6	2	1846	1639
7	2	1509	1473
8	2	822	902
9	2	439	525
13	2	741	497
14	2	569	740
15	2	365	531
17	2	674	724
22	2	414	291
0	3	2026	1904
1	3	2523	2743
2	3	1899	1930
3	3	3704	3225
4	3	2629	2137
5	3	808	474
6	3	3498	3228
7	3	602	580
10	3	3085	2991
12	3	1972	1955
13	3	659	665
14	3	535	566
15	3	656	792
16	3	1027	990
17	3	503	548
19	3	421	654
20	3	358	349
21	3	634	667
0	4	520	643
1	4	3974	4017
2	4	535	557
3	4	1870	1720
6	4	559	684
9	4	1314	1268
11	4	873	755
12	4	1104	1168
13	4	505	308
14	4	359	511
15	4	482	428
16	4	431	381
18	4	437	364
21	4	384	424
0	5	331	292
1	5	974	1141
2	5	1223	1161
3	5	952	1297
4	5	1760	1936
5	5	494	303
7	5	2061	1868
1	12	737	741
1	12	755	646
2	12	613	551
3	12	519	443
12	12	400	213
0	13	475	441

L = -3

H	K	FO	FC
1	0	2483	2541
3	0	447	455
7	0	3801	2973
11	0	1420	1697
13	0	1741	1575
10	6	765	680
11	6	559	571
12	6	414	336
13	6	786	706
14	6	459	287
20	6	405	462
0	7	329	622
1	7	479	578
2	7	2704	2842
4	7	2491	2557
5	7	717	651
7	7	428	443
8	7	620	612
10	7	387	338
11	7	773	767
12	7	565	432
15	7	607	546
17	7	375	390
18	7	416	309
0	8	310	317
1	8	704	700
3	8	411	331
6	8	741	630
7	8	653	493
9	8	1055	911
12	8	337	167
17	8	515	576
1	9	385	557
4	9	428	396
6	9	638	603
10	9	580	610
12	9	458	475
14	9	509	556
15	9	311	168
16	9	701	728
18	9	425	365
1	10	329	459
3	10	376	344
5	10	1078	954
7	10	1514	1417
9	10	484	579
9	10	753	679
5	11	398	224
7	11	363	370
10	11	379	153
0	12	737	741
1	12	755	646
2	12	613	551
3	12	519	443
12	12	400	213
0	13	475	441

Table 7. (Continued)

15	0	854	690	5	4	2653	2246	6	8	902	805	4	1	1022	1319	10	4	476	373	19	8	490	411	19	1	542	570
17	0	564	537	7	4	970	915	7	8	400	354	5	1	1634	1547	11	4	278	136	0	9	473	474	22	1	432	412
0	1	2836	2884	8	4	2819	2925	8	8	731	701	7	1	294	129	12	4	784	817	1	9	815	974	0	2	509	470
1	1	5918	5392	9	4	432	593	9	8	946	921	11	1	1971	2068	14	4	623	850	2	9	414	458	1	2	4363	3894
2	1	2581	2290	10	4	440	363	10	9	629	726	12	1	305	269	17	4	364	409	3	9	668	659	2	2	901	937
3	1	820	703	11	4	862	868	11	8	907	811	13	1	1012	1090	18	4	441	400	4	9	663	687	3	2	4173	4066
4	1	1626	1587	12	4	418	518	13	8	327	237	15	1	580	521	0	5	1056	1275	5	9	856	765	4	2	4569	4651
5	1	3836	3214	15	4	1290	1186	15	8	399	439	16	1	405	256	1	5	640	544	6	9	503	615	5	2	3809	3822
6	1	2717	2514	17	4	880	834	16	8	428	415	17	1	601	645	2	5	1207	1499	7	9	1150	1073	6	2	2721	2819
7	1	759	515	18	4	439	368	17	8	904	854	18	1	413	489	5	5	1225	1169	8	9	369	488	7	2	3687	3606
9	1	1040	928	20	4	408	501	19	8	496	492	21	1	453	326	7	5	1944	1865	9	9	969	957	8	2	811	852
10	1	1738	1854	0	5	1008	1100	1	9	297	300	22	1	364	252	8	5	664	556	10	9	974	998	9	2	830	831
11	1	2792	2643	1	5	1127	1246	2	9	692	503	24	1	293	153	9	5	1736	1607	12	9	769	832	10	2	1984	1932
12	1	585	587	2	5	765	800	4	9	891	763	0	2	1504	1538	12	5	458	471	16	9	752	628	11	2	504	422
13	1	2630	2613	3	5	899	1051	5	9	1225	1020	1	2	2140	2385	13	5	471	322	18	9	594	484	12	2	2471	2424
14	1	668	704	5	5	1753	1388	6	9	1022	842	3	2	846	735	14	5	1139	1120	0	10	590	682	13	2	465	459
15	1	1772	1908	6	5	2474	2333	7	9	542	614	4	2	3483	2905	16	5	620	504	5	10	612	513	14	2	289	306
16	1	826	849	7	5	2152	1691	8	9	1232	931	5	2	1566	1606	17	5	363	387	7	10	1005	879	16	2	915	800
17	1	858	1018	8	5	1336	1231	9	9	405	327	6	2	406	489	19	5	388	248	9	10	500	341	17	2	561	534
18	1	454	511	9	5	1299	1128	10	9	893	728	7	2	1983	1530	0	6	2542	3349	14	10	433	321	18	2	543	533
20	1	425	462	10	5	450	306	14	9	311	69	8	2	493	661	1	6	729	912	1	11	325	274	21	2	477	495
21	1	584	558	11	5	369	173	16	9	379	277	9	2	937	1004	2	6	2704	3042	5	11	405	530	0	3	1262	1123
22	1	602	639	12	5	1276	1352	18	9	350	265	10	2	282	87	3	6	483	685	6	11	356	292	1	3	835	761
1	2	616	776	14	5	1123	1142	0	10	406	478	11	2	876	931	6	6	3151	2825	13	11	318	194	2	3	1987	1589
3	2	3231	3012	17	5	456	381	2	10	564	741	12	2	581	729	7	6	554	436	2	12	502	462	3	3	2267	2298
5	2	3769	3506	19	5	424	406	6	10	572	622	13	2	439	564	8	6	2743	2521	9	12	582	536	4	3	744	563
7	2	1325	1330	1	6	1690	2019	8	10	355	293	14	2	829	872	10	6	857	740	10	12	370	371	5	3	1298	945
8	2	961	1191	2	6	323	599	9	10	540	535	17	2	590	554	11	6	849	671	11	12	464	487	6	3	1144	1270
9	2	605	622	3	6	1715	1939	11	10	753	727	21	2	651	663	12	6	1099	878	12	12	453	386	7	3	384	428
10	2	2200	2255	4	6	1343	1369	16	10	416	365	23	2	585	512	13	6	914	805	0	13	507	530	8	3	500	719
11	2	1442	1196	5	6	1506	1572	18	10	426	394	0	3	1130	1180	14	6	1146	1050	2	13	327	251	10	3	1324	1339
12	2	2286	2202	6	6	686	550	1	11	429	457	1	3	2616	2622	15	6	842	830					13	3	700	642
13	2	471	623	7	6	958	829	2	11	375	474	2	3	2301	2263	16	6	414	341		L = -5			14	3	521	491
14	2	357	266	8	6	998	899	6	11	440	372	3	3	4908	4767	19	6	347	247	H	K	FO	FC	15	3	466	447
16	2	1228	1257	9	6	1088	923	7	11	476	303	4	3	3249	2934	22	6	395	370	1	0	3257	3473	16	3	423	263
18	2	479	375	10	6	777	904	12	11	443	235	5	3	4854	4314	0	7	637	669	3	0	410	919	17	3	600	614
19	2	527	505	11	6	684	796	0	12	552	505	6	3	1868	1812	2	7	1446	1556	5	0	2524	2379	19	3	512	625
21	2	525	534	12	6	628	611	1	12	612	479	7	3	3293	3308	4	7	1573	1721	7	0	1218	1194	20	3	390	431
23	2	346	361	14	6	508	479	3	12	332	173	8	3	1041	903	6	7	327	303	9	0	2798	2592	0	4	1553	1531
0	3	2304	1957	19	6	342	309	7	12	462	382	9	3	2494	2443	7	7	565	436	15	0	1096	894	2	4	2734	2850
1	3	3041	2993	0	7	446	499	9	12	371	412	10	3	1132	1050	8	7	983	875	17	0	1089	1100	3	4	516	428
2	3	1635	1690	1	7	897	1240					11	3	1637	1583	9	7	387	317	19	0	326	162	4	4	3214	3219
3	3	3119	2470	2	7	1480	1515		L = -4			12	3	1926	2026	10	7	781	780	0	1	643	534	5	4	637	680
4	3	2903	2594	3	7	466	370	H	K	FO	FC	14	3	336	249	11	7	850	1003	1	1	1621	1210	6	4	4615	4642
6	3	2593	2530	5	7	2385	2230	0	0	873	261	16	3	1187	1222	12	7	333	406	2	1	1367	1472	8	4	4334	4248
7	3	1163	1017	6	7	638	616	2	0	1605	1665	17	3	699	599	13	7	805	973	3	1	1726	1852	10	4	2139	2101
8	3	2394	2227	7	7	1732	1487	4	0	524	616	18	3	426	433	14	7	362	430	4	1	1227	950	11	4	1092	1114
9	3	1010	827	8	7	401	372	6	0	5787	5779	19	3	571	576	16	7	326	316	5	1	2426	2333	14	4	424	348
10	3	934	1041	9	7	892	834	8	0	1087	1012	20	3	386	432	17	7	615	467	6	1	1032	1041	15	4	964	853
12	3	798	734	11	7	1313	1193	10	0	2907	2855	22	3	381	388	18	7	487	491	7	1	2068	1954	17	4	1115	1120
13	3	1326	1326	12	7	536	505	12	0	3521	3632	0	4	302	686	20	7	383	271	8	1	942	780	0	5	1218	1410
14	3	504	408	13	7	1369	1287	14	0	3117	2872	1	4	3788	3909	0	8	761	867	10	1	1593	1725	1	5	2895	3510
17	3	498	487	14	7	317	264	16	0	2048	2180	2	4	361	73	1	8	754	710	11	1	3126	2940	2	5	3832	4124
0	4	2723	2563	15	7	562	528	18	0	1090	1154	3	4	2361	2317	3	8	379	398	12	1	1589	1612	3	5	1045	1302
1	4	459	559	16	7	423	488	22	0	687	624	4	4	825	606	4	8	417	445	13	1	2009	1954	4	5	1208	1237
2	4	1848	1493	18	7	419	382	0	1	2284	2456	6	4	670	775	12	8	398	320	14	1	620	555	5	5	950	616
3	4	673	840	3	8	606	787	1	1	2146	2218	7	4	1524	1568	13	8	535	640	15	1	1024	993	6	5	1429	1151
4	4	1039	952	4	8	395	278	2	1	719	780	8	4	1037	999	14	8	334	356	16	1	563	790	7	5	2517	2444
5	4	628	626	5	8	607	684	3	1	1081	1136	9	4	744	731	17	8	508	517	17	1	491	580	8	5	1609	1649

Table 7. (Continued)

9	5	1664	1526	4	10	1119	1183	20	2	573	624	2	7	541	383		L = -7			0	4	733	748	12	8	369	453
10	5	1342	1397	5	10	660	724	21	2	488	448	3	7	371	374	H	K	FO	FC	1	4	1579	1422	14	8	418	639
11	5	561	403	6	10	1260	1217	23	2	419	448	4	7	691	630	3	0	897	807	2	4	1846	1997	0	9	736	879
12	5	1017	1043	8	10	760	780	1	3	1445	1320	5	7	634	545	5	0	2944	3005	3	4	552	639	1	9	390	351
13	5	589	493	10	10	560	524	2	3	2806	2566	6	7	351	13	7	0	1084	1000	4	4	2326	2572	2	9	911	1014
14	5	1245	1242	11	10	590	585	3	3	3247	3251	8	7	1060	1206	9	0	3203	3396	5	4	494	547	3	9	617	769
16	5	415	377	12	10	610	608	4	3	2731	2690	10	7	475	470	11	0	1603	1596	6	4	1784	1695	4	9	703	725
1	6	732	773	13	10	461	498	5	3	3780	3828	11	7	904	1028	13	0	746	711	8	4	2077	2173	5	9	708	844
2	6	862	824	1	11	512	473	6	3	1805	1607	12	7	639	739	15	0	417	367	9	4	1073	1047	6	9	450	599
3	6	1805	1874	3	11	347	300	7	3	3337	3536	13	7	820	939	17	0	813	629	10	4	825	865	8	9	423	450
4	6	704	766	7	11	364	330	8	3	1295	1223	14	7	909	958	19	0	787	769	11	4	680	806	12	9	453	428
5	6	1112	1102	10	11	524	504	9	3	2654	2511	16	7	457	506	0	1	1434	1234	12	4	343	283	14	9	562	518
6	6	653	606	12	11	537	484	10	3	378	44	17	7	571	501	1	1	1765	1720	13	4	733	792	15	9	396	259
7	6	915	997	0	12	357	426	11	3	2231	1845	18	7	381	373	3	1	773	838	14	4	610	623	17	9	415	213
8	6	756	856	7	12	414	453	12	3	1080	1052	19	7	407	364	4	1	853	799	15	4	425	413	2	10	362	262
9	6	535	514	9	12	663	555	13	3	1179	1086	0	8	840	1036	5	1	2815	2784	17	4	654	665	4	10	766	624
10	6	427	463	11	12	393	266	16	3	1049	1095	2	8	492	667	6	1	1477	1331	18	4	801	749	5	10	349	410
11	6	1148	1074	11	13	476	342	18	3	793	795	3	8	1101	1224	7	1	3174	3224	20	4	385	374	6	10	795	590
13	6	1553	1618	5	14	361	277	0	4	1385	1441	7	8	1021	1062	8	1	1472	1271	0	5	2076	2214	7	10	368	397
15	6	1013	961					1	4	1575	1546	9	8	797	753	11	1	1211	947	1	5	2076	2522	12	10	472	300
16	6	338	159		L = -6			2	4	1239	1270	10	8	324	239	12	1	1220	1204	2	5	3370	3736	13	10	347	347
19	6	334	255	H	K	FO	FC	3	4	3135	3268	13	8	490	301	13	1	704	684	3	5	1085	1040	0	11	518	561
0	7	570	593	0	0	5459	5282	4	4	622	403	1	9	641	622	14	1	442	408	4	5	2011	2191	1	11	544	536
1	7	2071	2649	2	0	4225	3831	5	4	543	462	3	9	952	828	18	1	388	483	5	5	771	824	3	11	345	317
2	7	652	639	6	0	4174	4308	6	4	1033	814	4	9	312	334	20	1	341	339	6	5	396	590	4	11	363	314
3	7	1886	2247	8	0	3776	3548	7	4	2808	2781	5	9	964	805	21	1	424	286	7	5	1159	1099	7	11	353	242
4	7	294	25	10	0	1157	1147	8	4	461	418	6	9	823	906	0	2	1100	1047	8	5	1610	1497	10	11	322	273
5	7	428	86	12	0	2797	2462	9	4	1191	1211	7	9	1307	1203	1	2	882	870	9	5	1634	1442	12	11	429	431
6	7	883	852	14	0	2138	2234	10	4	407	457	8	9	633	696	2	2	390	269	10	5	635	613	3	12	298	52
7	7	1111	1009	16	0	943	864	14	4	1139	1073	9	9	1081	994	3	2	3090	3078	11	5	432	365	7	12	336	252
8	7	512	708	18	0	407	515	16	4	523	366	10	9	924	947	4	2	1937	1994	13	5	958	812	10	12	392	334
9	7	1368	1128	22	0	400	423	17	4	378	477	12	9	1003	986	5	2	3267	2995	15	5	530	596	1	13	434	372
11	7	942	766	0	1	1883	1869	19	4	572	550	14	9	388	424	6	2	1165	1099	17	5	349	357	7	13	333	307
13	7	1006	764	1	1	630	784	0	5	792	804	18	9	439	444	7	2	1630	1595	22	5	377	348				
15	7	892	825	2	1	1654	1467	1	5	591	671	0	10	870	893	8	2	517	489	4	6	928	920		L = -8		
0	8	456	576	3	1	1077	1053	2	5	985	814	1	10	390	567	10	2	416	495	5	6	691	449	H	K	FO	FC
2	8	616	654	4	1	2355	2435	3	5	808	959	3	10	507	622	11	2	334	515	6	6	705	532	0	0	3075	3423
4	8	495	392	6	1	639	724	4	5	314	460	6	10	392	264	12	2	1070	1021	7	6	693	436	2	0	721	926
5	8	365	411	7	1	1055	970	6	5	435	732	13	10	444	466	13	2	796	844	8	6	578	390	4	0	1536	1819
6	8	586	556	8	1	941	1006	7	5	1331	1775	3	11	508	475	14	2	829	773	11	6	611	576	6	0	4819	4488
7	8	1225	1162	10	1	1361	1390	8	5	1292	1182	5	11	586	567	17	2	971	972	12	6	459	490	8	0	4419	4807
8	8	556	488	13	1	1252	1243	9	5	1179	1305	10	11	357	333	19	2	491	448	13	6	1842	1894	10	0	2174	2429
9	8	1374	1413	16	1	888	794	10	5	572	767	12	11	399	237	22	2	558	537	14	6	497	389	12	0	531	444
10	8	332	129	17	1	929	979	12	5	808	780	16	11	333	51	0	3	471	325	15	6	1395	1398	16	0	592	713
11	8	981	813	18	1	735	593	13	5	851	817	1	12	580	664	2	3	822	596	0	7	343	387	18	0	433	541
13	8	416	392	21	1	414	309	14	5	878	806	3	12	352	388	3	3	1922	1779	1	7	2574	2958	0	1	908	984
15	8	507	450	0	2	849	903	16	5	700	628	6	12	349	318	4	3	2200	2465	3	7	1808	2172	1	1	699	713
16	8	413	324	1	2	886	903	0	6	2979	3401	9	12	339	278	5	3	2084	2288	5	7	1027	1140	2	1	1486	1422
17	8	644	650	2	2	4147	4164	1	6	1689	1867	11	12	403	372	6	3	1198	1602	7	7	743	623	3	1	608	644
19	8	445	488	3	2	974	1165	2	6	4308	4837	13	12	362	206	7	3	1676	1580	8	7	906	894	6	1	1067	1045
0	9	454	604	4	2	2323	2124	3	6	1185	1335	14	12	329	94	8	3	1859	2352	9	7	337	75	7	1	509	399
1	9	299	178	5	2	448	142	4	6	2322	2849	0	13	402	333	9	3	1106	1110	11	7	493	588	8	1	919	989
3	9	302	175	7	2	724	477	5	6	833	1076	4	13	358	233	10	3	2115	2301	2	8	925	1050	9	1	1683	1841
5	9	361	155	9	2	671	604	6	6	973	908	10	13	420	416	12	3	307	306	4	8	347	364	10	1	2172	2233
7	9	591	651	11	2	1365	1423	8	6	1733	1532	12	13	409	205	14	3	414	26	7	8	552	559	12	1	1123	1007
14	9	371	404	12	2	752	543	10	6	656	540	6	14	284	118	16	3	816	614	8	8	494	483	13	1	533	459
18	9	518	372	13	2	599	769	13	6	699	768					19	3	726	942	9	8	950	991	15	1	368	289
0	10	681	723	14	2	502	428	15	6	720	692					20	3	365	212	10	8	341	355	17	1	640	669
2	10	740	809	18	2	682	705	0	7	457	325					24	3	318	119	11	8	762	843	19	1	435	449

Table 7. (Continued)

H	K	FO	FC
0	2	908	743
1	2	1301	913
3	2	1597	1908
4	2	733	814
5	2	1361	1573
6	2	580	634
7	2	689	774
8	2	477	223
9	2	663	481
10	2	812	610
11	2	474	521
12	2	419	312
15	2	492	601
16	2	390	532
18	2	798	787
20	2	428	640
0	3	983	1133
2	3	1383	1564
4	3	1512	1498
5	3	635	812
7	3	1713	1595
8	3	647	549
9	3	348	253
10	3	561	556
11	3	964	845
12	3	1133	959
13	3	746	815
14	3	652	679
15	3	856	748
17	3	790	844
18	3	1136	1236
19	3	871	861
20	3	400	307
21	3	673	641
0	4	1602	1839
1	4	536	367
2	4	335	234
3	4	1092	831
4	4	1687	1584
6	4	1690	2006
7	4	1801	1960
8	4	764	867
9	4	1662	1848
11	4	829	961
13	4	636	612
14	4	863	854
15	4	697	667
16	4	450	276
0	5	316	391
1	5	277	490
3	5	409	380
5	5	787	666
6	5	803	884
7	5	481	445
8	5	399	650
9	5	836	810
11	5	357	315
13	5	1132	1126
14	5	575	562
15	5	665	719
16	5	356	294
19	5	459	539
0	6	1873	2069
1	6	487	605
2	6	2664	3096
3	6	1417	1272
4	6	2374	2432
5	6	709	867
7	6	433	418
8	6	675	708
9	6	574	563
11	6	379	330
12	6	1237	1400
13	6	349	276
14	6	1147	1156
16	6	313	323
0	7	617	808
3	7	914	968
4	7	480	256
5	7	668	682
6	7	612	456
7	7	345	347
10	7	427	524
11	7	343	273
12	7	559	541
13	7	415	462
14	7	692	774
16	7	473	470
0	8	421	532
1	8	338	357
3	8	531	555
6	8	445	519
7	8	656	609
9	8	422	461
10	8	497	509
12	8	459	531
14	8	371	350
15	8	463	423
16	8	346	228
1	9	374	246
2	9	543	651
3	9	405	387
6	9	367	453
7	9	358	217
8	9	383	289
9	9	392	318
10	9	388	313
12	9	756	611
14	9	733	617
17	9	630	417
0	10	762	789
1	10	513	612
3	10	942	929
5	10	834	981
11	10	380	236
14	10	401	271
16	10	395	265
2	11	316	354
5	11	444	278
9	11	514	462
11	11	480	427
0	12	325	304
1	12	522	617
5	12	335	179
8	12	504	423
10	13	469	375

L = -9

H	K	FO	FC
1	0	2871	2641
3	0	2801	2667
7	0	2451	2434
9	0	2789	2723
11	0	2255	2327
15	0	796	903
17	0	427	263
19	0	619	501
0	1	1393	1300
1	1	839	472
2	1	942	936
3	1	964	855
4	1	1399	1497
7	1	1479	1814
8	1	2045	1973
9	1	2719	2800
10	1	1596	1588
11	1	1062	1242
13	1	301	336
15	1	703	752
16	1	416	401
17	1	704	808
19	1	415	450
0	2	1018	990
1	2	543	651
2	2	1206	1417
3	2	1107	1030
4	2	1143	1148
5	2	676	760
6	2	1006	1144
7	2	1231	1170
8	2	1233	1352
9	2	887	853
10	2	1223	1345
11	2	808	829
13	2	428	361
14	2	1215	1253
15	2	583	701
16	2	485	380
17	2	1034	1060
19	2	854	870
21	2	354	335
22	2	462	401
0	3	297	493
1	3	1416	1396
2	3	1511	1840
3	3	1029	984
4	3	1432	1732
5	3	1249	1290
6	3	2569	2765
8	3	1863	1978
9	3	1442	1248
10	3	550	643
14	3	386	295
16	3	565	449
19	3	563	645
21	3	411	416
0	4	1824	1894
1	4	403	591
2	4	591	514
3	4	1985	2121
5	4	537	428
6	4	1396	1316
7	4	545	495
8	4	980	1205
9	4	828	677
12	4	420	669
13	4	569	611
14	4	359	351
15	4	634	533
16	4	340	191
17	4	363	320
18	4	1005	906
20	4	755	754
0	5	379	113
1	5	1354	1431
2	5	806	665
3	5	1022	1140
4	5	1674	1669
5	5	451	394
6	5	717	719
7	5	1196	1052
8	5	1036	1170
10	5	298	190
11	5	467	431
12	5	1379	1314
13	5	1385	1460
14	5	972	953
15	5	989	974
16	5	421	403
17	5	455	321
0	6	505	512
1	6	391	480
3	6	414	588
4	6	530	447
5	6	375	135
6	6	1213	1172
7	6	305	10
8	6	617	651
9	6	456	404
13	6	979	928
14	6	495	556
15	6	950	835
0	7	548	588
1	7	540	495
3	7	1028	1078
4	7	785	850
5	7	973	952
6	7	363	313
8	7	538	644
10	7	556	579
11	7	1344	1522
13	7	1346	1397
15	7	687	733
0	8	295	419
2	8	442	459
4	8	410	413
5	8	872	966
7	8	317	196
14	8	418	456
18	8	468	455
0	9	881	1015
2	9	1206	1216
3	9	854	990
4	9	573	590
5	9	1324	1438
6	9	478	299
7	9	477	416
8	9	433	471
3	10	338	424
14	10	395	326
16	10	583	446
1	11	384	334
4	11	419	360
1	12	324	298
10	12	448	486
12	12	479	392
1	13	345	216
7	13	444	311
9	13	433	273

L =-10

H	K	FO	FC
0	0	844	623
2	0	690	555
4	0	553	298
6	0	1321	1546
8	0	3747	3949
10	0	3769	3758
12	0	1311	1587
14	0	882	987
16	0	1303	1340
18	0	863	864
0	1	1350	1414
1	1	1397	1365
3	1	1302	1183
7	1	384	438
8	1	1638	1618
9	1	443	671
10	1	2531	2559
11	1	659	847
12	1	916	836
13	1	330	327
14	1	547	702
15	1	552	525
16	1	843	1027
17	1	297	26
18	1	587	545
19	1	565	569
0	2	1780	1491
1	2	571	726
2	2	884	896
3	2	1847	1938
4	2	322	125
5	2	2593	2793
6	2	707	778
8	2	1005	923
9	2	1151	1214
10	2	791	853
11	2	606	586
18	2	396	431
19	2	474	390
20	2	352	314
21	2	542	508
0	3	2272	2245
1	3	2013	2078
2	3	755	859
3	3	2231	2451
4	3	946	1162
5	3	1845	2156
7	3	1940	2098
8	3	841	715
9	3	1317	1390
10	3	1357	1325
11	3	920	1109
12	3	798	669
14	3	666	654
15	3	607	489
17	3	901	865
18	3	937	906
19	3	741	607
20	3	552	585
21	3	491	481
0	4	1405	1505
1	4	897	1009
3	4	451	238
4	4	1516	1633
6	4	1477	1522
7	4	993	1295
9	4	2095	2089
10	4	312	485
11	4	295	390
13	4	1224	1068
15	4	602	505
16	4	380	295
18	4	328	188
2	5	358	615
7	5	1138	1261
9	5	594	592
10	5	428	520
13	5	693	803
15	5	633	757
19	5	449	484
2	6	390	437
4	6	922	814
5	6	412	459
6	6	834	741
9	6	582	505
10	6	509	475
11	6	748	685
12	6	1691	1844
13	6	820	894
14	6	1679	1732
15	6	488	565
16	6	678	673
18	6	318	123
0	7	546	640
2	7	385	347
3	7	756	805
5	7	861	881
6	7	527	373
7	7	335	388
8	7	372	244
10	7	550	697
14	7	404	466
1	8	668	830
2	8	662	821
4	8	887	1046
5	8	489	354
6	8	787	803
10	8	495	572
12	8	652	622
17	8	385	330
1	9	383	341
2	9	645	838
3	9	395	368
4	9	964	1087
5	9	565	683
6	9	538	519
7	9	813	848
9	9	522	409
15	9	352	153
17	9	501	366
1	10	358	343
3	10	987	978
5	10	731	689
6	10	473	708
14	10	409	265
16	10	444	272
2	11	323	295
9	11	598	585
11	11	391	463
12	11	345	277
16	11	393	260
7	12	391	326
8	12	473	395
9	12	382	424
10	12	521	450
11	12	359	277
2	13	371	332
4	13	346	275
6	13	370	260

Table 7. (Continued)

L =-11

H	K	FO	FC
1	0	3255	3396
5	0	2229	1938
7	0	361	374
9	0	1356	1363
11	0	1109	1204
13	0	507	544
15	0	1389	1478
17	0	809	826
0	1	1193	1296
1	1	3145	2980
2	1	444	433
3	1	2146	2062
4	1	1751	1935
5	1	1331	1228
6	1	740	746
7	1	1153	1233
8	1	1339	1425
9	1	1865	2132
10	1	1934	1807
11	1	1428	1420
12	1	1059	1028
15	1	582	525
17	1	435	507
19	1	348	426
0	2	925	856
1	2	1062	1152
2	2	838	828
3	2	404	164
4	2	2684	2700
5	2	546	342
6	2	2464	2603
7	2	1358	1378
8	2	813	890
9	2	2259	2172
10	2	1507	1744
11	2	849	795
13	2	336	276
14	2	852	793
16	2	643	750
17	2	742	842
19	2	669	734
0	3	855	1002
1	3	1975	2152
2	3	695	903
3	3	422	328
4	3	1377	1618
5	3	1534	1416
6	3	1041	1161
7	3	511	598
8	3	379	484
9	3	519	638
11	3	1281	1479
17	3	322	46
18	3	475	487
20	3	498	444
22	3	551	459
0	4	1859	1938
2	4	1874	1819
3	4	1348	1509
4	4	1488	1965
5	4	782	1012
6	4	2411	2523
7	4	570	434
8	4	1807	2138
10	4	1024	1233
13	4	482	446
14	4	336	435
15	4	428	450
18	4	643	449
20	4	584	623
23	4	400	212
0	5	1520	1740
2	5	809	960
3	5	492	694
4	5	393	351
5	5	273	133
6	5	714	653
7	5	562	685
10	5	380	372
11	5	572	662
12	5	1071	1051
13	5	862	958
14	5	1287	1360
15	5	888	888
16	5	380	496
19	5	355	307
0	6	703	712
2	6	401	309
3	6	470	434
4	6	466	563
5	6	297	220
6	6	1005	990
8	6	605	723
11	6	822	834
15	6	431	408
0	7	280	302
1	7	1077	1276
2	7	307	250
3	7	573	706
4	7	857	778
5	7	288	235
6	7	565	551
9	7	568	611
10	7	422	458
11	7	1124	1162
13	7	1458	1552
15	7	618	741
0	8	812	839
3	8	1059	1036
4	8	308	386
5	8	1181	1345
7	8	1055	1015
8	8	393	273
9	8	393	354
15	8	352	225
0	9	523	534
2	9	808	809
3	9	808	851
5	9	1179	1205
7	9	704	697
9	9	428	458
18	9	348	310
2	10	466	518
3	10	435	608
4	10	877	914
5	10	403	423
6	10	633	842
8	10	784	826
10	10	613	561
16	10	504	317
4	11	397	284
8	11	437	432
9	11	314	246
10	11	561	573
1	12	507	490
10	12	359	351
12	12	359	325
7	13	346	93
9	13	416	233

L =-12

H	K	FO	FC
0	0	2698	2574
2	0	2543	2462
4	0	2648	2649
6	0	517	449
8	0	1788	1857
10	0	3043	3235
12	0	1134	1200
16	0	353	372
18	0	552	503
22	0	393	64
0	1	908	1045
1	1	1482	1499
2	1	1366	1465
3	1	452	476
4	1	246	25
5	1	799	679
6	1	906	747
8	1	913	823
9	1	321	250
10	1	736	762
11	1	292	225
12	1	329	148
13	1	312	192
14	1	1007	1042
15	1	857	977
16	1	1422	1447
18	1	438	411
19	1	392	444
0	2	1146	1205
1	2	769	941
2	2	982	961
3	2	1179	1288
5	2	1271	1238
6	2	1240	1189
7	2	919	1036
8	2	435	395
9	2	1741	1894
10	2	287	359
11	2	1329	1270
15	2	337	219
19	2	404	204
21	2	442	493
23	2	309	113
0	3	1817	1903
1	3	1050	1163
3	3	2576	2558
4	3	545	719
5	3	2494	2686
6	3	511	348
7	3	2668	2812
8	3	1058	1030
9	3	1835	1936
10	3	1392	1278
11	3	1307	1264
12	3	346	213
13	3	542	560
18	3	530	488
20	3	387	448
24	3	348	287
0	4	995	1138
1	4	674	821
2	4	828	970
5	4	398	297
6	4	1046	963
7	4	359	204
8	4	301	36
9	4	532	451
10	4	620	728
13	4	608	524
15	4	479	378
19	4	576	518
0	5	406	421
1	5	584	665
2	5	561	693
3	5	375	360
4	5	625	729
7	5	857	1034
8	5	272	195
10	5	631	634
11	5	508	415
15	5	352	379
20	5	464	404
0	6	977	911
1	6	1027	1139
2	6	1102	1240
3	6	705	713
4	6	344	193
5	6	435	446
6	6	412	371
8	6	325	196
11	6	548	514
12	6	907	999
13	6	656	644
14	6	1195	1148
16	6	539	526
1	7	568	575
5	7	636	683
6	7	466	478
7	7	440	549
9	7	320	366
10	7	729	854
12	7	781	802
0	8	428	431
1	8	763	900
2	8	676	824
4	8	911	1069
5	8	670	581
6	8	820	820
7	8	935	797
12	8	462	574
1	9	564	679
2	9	563	682
3	9	531	579
4	9	1022	1028
5	9	849	875
6	9	1045	1041
7	9	966	981
8	9	529	543
9	9	577	473
12	9	364	396
6	10	605	801
8	10	357	353
9	10	373	574
11	10	416	502
5	11	319	238
12	11	333	248
15	11	372	138
8	12	406	304
5	13	336	143

L =-13

H	K	FC	FC
1	0	1615	1742
3	0	962	741
5	0	619	463
7	0	503	286
13	0	532	631
15	0	1541	1580
17	0	1124	1212
21	0	447	542
23	0	375	248
0	1	830	916
1	1	2972	2862
2	1	727	820
3	1	1540	1627
4	1	1341	1355
5	1	935	892
6	1	1266	1332
8	1	552	483
9	1	1034	988
10	1	1107	1166
11	1	490	587
12	1	694	912
15	1	621	604
0	2	1274	1281
1	2	1222	1181
4	2	2198	2112
6	2	2052	2066
7	2	1237	1184
1	7	1206	1247
3	7	1331	1507
4	7	346	413
5	7	516	586
6	7	589	471
8	7	418	308
13	7	646	658
0	8	606	723
2	8	372	517
3	8	853	866
5	8	1159	1228
7	8	701	603
8	8	332	354
5	9	589	612
6	9	597	477
7	9	500	558
8	9	645	566
10	9	465	352
16	9	373	334
18	9	364	249
3	10	357	308
4	10	609	551
6	10	738	676
8	10	684	596
10	10	638	521
0	11	389	220
10	11	452	503
12	11	341	315
1	12	352	342

L =-14

H	K	FO	FC
0	0	3199	3156
2	0	2647	2713
4	0	2253	2335
6	0	1985	1873
8	0	569	422
14	0	1268	1441
16	0	800	772
20	0	451	442
0	1	1214	1340
1	1	787	901
2	1	1426	1602
5	1	918	919
7	1	556	455
8	1	539	474
9	1	871	792
14	1	753	843
15	1	438	426
16	1	482	652
20	1	557	469
5	6	326	261
6	6	431	292
7	6	412	436
9	6	894	887
11	6	1036	1009
12	6	602	637
13	6	516	556
14	6	336	152
1	7	1206	1247
3	7	1331	1507
4	7	346	413
5	7	516	586
6	7	589	471
8	7	418	308
13	7	646	658
0	8	606	723
2	8	372	517
3	8	853	866
5	8	1159	1228
7	8	701	603
8	8	332	354
5	9	589	612
6	9	597	477
7	9	500	558
8	9	645	566
10	9	465	352
16	9	373	334
18	9	364	249
3	10	357	308
4	10	609	551
6	10	738	676
8	10	684	596
10	10	638	521
0	11	389	220
10	11	452	503
12	11	341	315
1	12	352	342

Table 7. (Continued)

H	K	FO	FC
0	2	538	563
1	2	440	548
2	2	562	612
4	2	707	762
5	2	737	951
6	2	1142	1232
7	2	393	526
8	2	637	653
9	2	538	561
11	2	787	797
12	2	874	954
14	2	549	702
15	2	501	559
17	2	338	400
18	2	343	376
2	3	782	847
2	3	432	491
3	3	610	676
4	3	847	836
5	3	1181	1164
6	3	926	824
7	3	851	950
8	3	624	841
9	3	502	678
10	3	625	705
11	3	705	659
13	3	366	349
15	3	479	355
17	3	675	559
19	3	504	565
21	3	429	376
0	4	288	516
2	4	1001	1153
3	4	367	305
5	4	698	743
6	4	789	644
7	4	780	854
8	4	526	361
11	4	475	440
0	5	366	384
1	5	354	446
2	5	612	652
8	5	719	787
10	5	457	475
11	5	614	540
12	5	385	372
13	5	802	777
16	5	376	397
17	5	428	532
0	6	1103	1006
1	6	672	687
2	6	962	934
3	6	977	1007
4	6	654	623
5	6	718	524
6	6	662	530
7	6	453	356
8	6	756	545
9	6	321	190
10	6	744	632
12	6	576	482
1	7	930	1080
2	7	519	669
5	7	451	331
7	7	544	530
10	7	747	652
12	7	941	858
14	7	416	397
15	7	382	251
4	8	372	457
5	8	562	352
6	8	561	641
7	8	628	624
16	8	352	480
18	8	506	440
0	9	352	433
4	9	602	616
6	9	692	627
7	9	379	287
9	9	459	441
11	9	458	270
6	10	345	297
8	10	358	480
9	10	544	544
1	12	435	292
5	12	290	94

L =-15

H	K	FO	FC
1	0	375	524
3	0	1269	1340
7	0	415	493
9	0	1067	967
11	0	977	941
13	0	350	481
15	0	1093	1129
17	0	919	981
21	0	446	471
23	0	411	333
0	1	322	337
1	1	638	589
4	1	380	526
6	1	600	718
8	1	1041	1146
9	1	411	389
10	1	364	400
11	1	314	290
12	1	386	294
13	1	1092	1077
14	1	483	498
15	1	1051	1078
16	1	527	480
17	1	535	508
0	2	709	775
1	2	381	270
2	2	1033	996
3	2	618	635
5	2	985	923
7	2	400	282
10	2	523	589
11	2	465	569
14	2	351	254
20	2	433	527
22	2	417	413
0	3	957	830
2	3	1352	1256
4	3	1468	1469
5	3	698	725
6	3	1008	1094
7	3	1458	1557
11	3	1082	1140
13	3	550	670
2	4	474	440
3	4	308	152
4	4	721	733
5	4	740	779
6	4	880	969
7	4	303	148
8	4	615	506
10	4	326	415
13	4	366	428
16	4	604	584
18	4	614	574
1	5	292	427
2	5	359	302
9	5	578	374
10	5	694	752
12	5	898	900
20	5	335	179
1	6	1075	1123
3	6	725	796
5	6	435	333
7	6	729	941
9	6	825	887
10	6	692	694
11	6	973	943
12	6	805	746
13	6	708	632
14	6	561	612
17	6	377	403
3	7	711	689
5	7	561	482
6	7	336	318
7	7	322	257
9	7	439	501
11	7	431	543
12	7	357	481
13	7	374	391
0	8	462	567
2	8	430	593
5	8	319	240
17	8	361	292
1	9	397	377
3	9	509	321
6	9	723	481
8	9	776	645
10	9	479	313
12	10	298	138

L =-16

H	K	FO	FC
0	0	1035	986
2	0	828	809
4	0	335	448
6	0	878	767
8	0	1171	1009
10	0	918	899
12	0	1202	1088
14	0	1704	1436
16	0	1334	1291
20	0	456	415
22	0	372	215
0	1	290	337
1	1	755	679
2	1	910	998
4	1	694	712
5	1	381	328
6	1	719	677
7	1	499	518
8	1	433	413
9	1	458	505
10	1	444	408
11	1	574	472
12	1	427	355
1	2	730	443
2	2	410	519
3	2	1046	962
4	2	780	803
5	2	488	379
6	2	1071	1203
7	2	274	325
8	2	634	768
12	2	1081	1137
14	2	922	782
15	2	577	646
1	3	961	949
3	3	450	566
4	3	799	692
5	3	994	952
6	3	1186	1113
7	3	910	892
8	3	625	522
9	3	887	836
11	3	572	464
15	3	531	621
17	3	567	564
19	3	543	492
21	3	485	421
0	4	389	379
1	4	943	782
2	4	449	535
3	4	1064	1068
4	4	365	362
5	4	1088	1109
6	4	616	511
7	4	941	983
8	4	683	696
11	4	645	792
12	4	640	710
13	4	583	678
0	5	337	239
1	5	670	580
2	5	575	413
3	5	545	430
5	5	365	476
6	5	576	827
7	5	877	993
8	5	496	687
9	5	655	610
10	5	514	546
11	5	640	570
13	5	640	705
14	5	385	353
16	5	363	471
17	5	386	378
1	6	337	55
4	6	553	488
5	6	526	395
6	6	580	546
8	6	592	459
9	6	524	468
10	6	1254	1135
12	6	1157	1057
14	6	456	339
17	6	328	201
0	7	367	286
1	7	689	869
2	7	695	784
3	7	374	539
4	7	304	341
8	7	469	447
10	7	527	415
12	7	583	427
18	7	506	370
0	8	447	506
2	8	331	412
7	8	438	342
18	8	456	380
2	9	367	370
5	9	408	306
7	9	347	255
1	10	330	266
7	10	455	220
6	12	319	160

L =-17

H	K	FC	FC
1	0	349	214
3	0	1163	1427
5	0	444	484
0	1	409	302
2	1	364	321
3	1	675	863
5	1	293	407
6	1	562	615
8	1	763	888
9	1	556	511
11	1	826	700
13	1	918	832
15	1	1019	960
16	1	396	447
17	1	611	496
0	2	416	246
2	2	1160	1103
3	2	821	821
4	2	996	914
5	2	1334	1282
6	2	285	83
7	2	1212	1128
8	2	368	311
9	2	387	405
10	2	598	451
16	2	403	291
0	3	828	769
2	3	1236	1108
4	3	1313	1253
5	3	677	765
6	3	672	603
7	3	852	966
8	3	387	102
9	3	496	569
11	3	513	448
13	3	691	676
20	3	343	301
2	4	333	362
4	4	778	801
6	4	1300	1317
8	4	1346	1373
9	4	617	685
10	4	660	629
15	4	602	498
16	4	488	430
18	4	619	619
0	5	721	758
2	5	523	539
3	5	559	498
7	5	332	256
9	5	550	361
10	5	597	590
11	5	583	418
12	5	998	1021
13	5	444	313
14	5	529	545
1	6	1118	951
3	6	827	780
7	6	580	795
9	6	522	649
10	6	336	391
12	6	507	588
14	6	468	473
17	6	372	426
0	7	354	238
1	7	594	698
7	7	375	397
10	7	379	247
11	7	760	640
12	7	351	295
13	7	789	738
16	7	374	232
1	8	350	353
2	8	328	479
9	8	343	290
11	8	424	316
1	9	585	421
3	9	452	308
5	9	334	178
8	9	377	363
10	9	319	156
13	9	364	235
2	10	506	511
4	10	493	509
8	10	329	265

L =-18

H	K	FO	FC
0	0	757	841
2	0	437	443
4	0	363	541
8	0	1040	1020
10	0	1268	1175
12	0	831	724
14	0	854	734
16	0	659	500
18	0	331	251
1	1	404	318
2	1	675	640
3	1	606	523
4	1	937	1059
6	1	541	526
7	1	375	410
14	1	594	483
1	2	708	708
3	2	726	631
5	2	525	492
6	2	581	839
8	2	382	435
9	2	647	832
10	2	465	261
12	2	605	593
13	2	324	127
14	2	359	383
18	2	340	401
1	3	689	552
3	3	1340	1286
5	3	1517	1505
6	3	1165	1037
7	3	1538	1408
8	3	670	687
9	3	1266	1191
11	3	653	638
12	3	326	255
15	3	326	344

Table 7. (Continued)

3 4 891 952	13 1 352 294	10 1 747 572	5 6 581 428
5 4 740 657	16 1 327 205	12 1 528 405	6 6 328 252
7 4 365 361	18 1 318 222	13 1 291 35	10 6 353 202
8 4 499 640	2 2 1007 937	3 2 317 284	1 7 341 309
11 4 519 726	3 2 444 504	9 2 447 394	14 7 324 234
12 4 512 500	4 2 918 702	12 2 336 87	16 7 362 135
13 4 418 574	5 2 720 827	13 2 451 431	6 9 338 235
15 4 302 67	6 2 563 445	16 2 417 325	10 9 297 139
17 4 477 416	7 2 912 954	18 2 454 437	
0 5 362 392	9 2 452 466	3 3 520 508	L =-22
1 5 486 404	10 2 550 254	5 3 505 606	H K FO FC
2 5 388 365	12 2 482 199	6 3 397 385	6 0 391 341
3 5 499 510	20 2 306 159	7 3 462 399	8 0 619 513
5 5 375 352	0 3 454 442	8 3 462 404	10 0 724 555
7 5 623 678	2 3 564 504	9 3 620 435	14 0 661 663
8 5 480 520	4 3 489 529	1 4 340 279	16 0 331 312
10 5 309 333	7 3 723 697	14 4 372 391	3 1 406 346
14 5 520 435	9 3 547 533	17 4 507 453	6 1 379 261
16 5 464 398	13 3 710 656	18 4 324 42	8 1 828 741
17 5 375 194	14 3 517 377	2 5 354 410	9 1 531 554
0 6 596 640	16 3 391 305	3 5 400 330	10 1 780 733
1 6 483 482	18 3 442 382	4 5 407 307	18 1 375 229
2 6 471 577	1 4 344 151	5 5 337 421	3 2 487 304
8 6 412 381	5 4 364 374	0 6 517 508	13 2 454 391
10 6 721 631	6 4 913 959	1 6 651 556	15 2 442 507
11 6 362 370	8 4 999 1025	2 6 612 577	18 2 340 228
12 6 735 710	10 4 538 484	3 6 426 344	5 3 339 390
14 6 599 452	12 4 333 304	10 6 330 180	12 3 365 175
1 7 356 458	14 4 324 151	4 7 441 341	17 3 402 286
2 7 581 533	0 5 523 551	6 7 334 209	18 3 368 139
18 7 393 357	1 5 337 388	8 8 336 91	19 3 455 308
0 8 666 684	2 5 773 767	4 9 360 274	1 4 394 374
2 8 356 484	3 5 319 280		14 4 449 279
13 8 333 396	4 5 423 389	L =-21	4 5 489 486
2 9 368 389	12 5 427 472	H K FO FC	5 5 567 576
3 9 519 386	14 5 363 458	7 0 864 789	7 5 430 411
4 9 449 476	1 6 516 411	9 0 1105 1060	13 5 360 153
5 9 568 479	3 6 769 685	11 0 935 768	2 6 361 244
10 9 430 379	6 6 297 171	15 0 408 317	10 6 478 425
5 11 366 327	1 7 709 770	0 1 419 521	6 7 462 315
	3 7 439 505	2 1 302 138	6 8 382 285
L =-19	6 8 433 239	7 1 537 509	
H K FO FC	12 9 295 38	14 1 410 424	L =-23
3 0 855 958	4 10 551 511	1 2 306 190	H K FC FC
5 0 345 386	6 10 462 393	8 2 334 413	7 0 499 490
7 0 625 475		11 2 394 179	9 0 905 856
9 0 780 679	L =-20	17 2 370 289	11 0 610 656
11 0 411 259	H K FO FC	4 3 358 255	1 1 312 290
13 0 528 366	0 0 893 1059	5 3 324 226	3 1 330 323
15 0 370 227	2 0 491 576	8 3 352 188	9 1 403 275
0 1 613 749	4 0 790 871	9 3 350 253	11 1 406 257
1 1 723 728	6 0 391 440	10 3 328 206	13 1 351 358
2 1 508 390	12 0 328 238	14 3 448 307	15 1 333 316
3 1 474 538	0 1 603 501	18 3 375 250	3 2 404 305
4 1 446 342	3 1 474 429	19 3 438 342	11 2 360 238
5 1 524 603	4 1 451 577	2 4 393 404	0 3 451 365
8 1 421 405	7 1 453 560	4 4 410 421	2 3 472 495
10 1 356 282	8 1 731 599	15 5 359 89	4 3 379 339
11 1 545 518	9 1 476 545	3 6 469 536	6 3 397 295

3 4 311 227			
4 4 497 581			
6 4 424 591			
0 5 352 338			
9 5 312 137			
11 7 436 480			
L =-24			
H K FC FC			
6 0 336 70			
8 0 500 486			
10 0 383 302			
14 0 453 332			
8 1 332 418			
2 2 385 414			
5 2 426 348			
3 3 528 531			
4 3 365 322			
5 3 586 555			
7 3 612 518			
1 4 556 455			
16 4 347 43			
5 5 380 410			
7 5 353 344			
10 6 387 330			
12 6 545 401			
8 7 331 124			
L =-25			
H K FC FC			
11 0 347 273			
3 1 381 356			
4 1 359 230			
7 2 348 328			
12 2 363 24			
1 3 437 324			
4 4 492 372			
6 4 442 501			
12 5 353 194			
13 5 350 45			
L =-26			
H K FC FC			
12 2 329 132			
L =-27			
H K FC FC			
5 0 462 311			
7 0 449 277			

Table 8. Final parameters[a] for $(NaC^+)_3Sb_7^{3-}$

Positional parameters for heavy atoms

	x	y	z
Sb(1)	0.22558(15)	0.18851(22)	0.41768(10)
Sb(2)	0.17610(12)	0.07328(20)	0.47838(10)
Sb(3)	0.32617(13)	0.25736(22)	0.50100(12)
Sb(4)	0.37464(11)	0.09264(20)	0.55983(10)
Sb(5)	0.26285(14)	0.33736(20)	0.56892(13)
Sb(6)	0.27292(12)	0.01810(20)	0.57388(10)
Sb(7)	0.19929(12)	0.18028(21)	0.58058(10)
Na(1)	0.66271(59)	0.28473(89)	0.61846(50)
Na(2)	0.53016(61)	0.3711(10)	0.28429(51)
Na(3)	0.57729(59)	0.8286(11)	0.13623(53)

Anisotropic thermal parameters ($\times 10^3$)[b] for heavy atoms

	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
Sb(1)	5.9(1)	12.7(2)	2.22(5)	3.9(1)	0.36(6)	0.34(9)
Sb(2)	3.27(7)	11.3(2)	3.26(6)	0.4(1)	-0.14(5)	-1.88(9)
Sb(3)	3.91(8)	11.6(2)	4.46(8)	0.3(1)	1.39(6)	2.2(1)
Sb(4)	2.68(6)	10.9(2)	3.64(6)	1.8(1)	0.57(5)	0.7(1)
Sb(5)	4.50(9)	8.5(2)	5.32(9)	0.5(1)	1.15(7)	-1.9(1)
Sb(6)	3.95(8)	10.2(2)	3.05(6)	0.5(1)	0.55(5)	0.92(9)
Sb(7)	3.78(8)	11.3(2)	3.43(6)	1.0(1)	1.57(6)	-0.2(1)
Na(1)	3.8(3)	6.8(8)	2.8(3)	2.0(4)	1.0(2)	0.7(4)
Na(2)	3.5(3)	12(1)	2.8(3)	2.0(5)	1.1(2)	1.0(4)
Na(3)	3.0(3)	13(1)	3.1(3)	-1.6(6)	0.8(2)	0.1(5)

[a]Estimated standard deviations in parameters from ORFLS[60] are in parentheses.

[b]The anisotropic temperature factors have the general form $\exp-(h^2\beta_{11} + k^2\beta_{22} + \ell^2\beta_{33} + 2hk\beta_{12} + 2h\ell\beta_{13} + 2k\ell\beta_{23})$.

Table 8. (Continued)

Positional and thermal parameters for the first crypt

	x	y	z	B
N(101)	0.6057(14)	0.0948(22)	0.6186(12)	8.8(8)
C(102)	0.6020(21)	0.0839(34)	0.6771(18)	10.6(13)
C(103)	0.5829(19)	0.1808(32)	0.6954(16)	9.9(11)
O(104)	0.6282(11)	0.2516(17)	0.7031(9)	8.6(6)
C(105)	0.6810(21)	0.2450(34)	0.7536(18)	10.6(12)
C(106)	0.7205(21)	0.3245(35)	0.7619(19)	10.9(13)
O(107)	0.7373(12)	0.3329(20)	0.7122(11)	10.4(7)
C(108)	0.7718(23)	0.4239(39)	0.7184(21)	12.8(15)
C(109)	0.7704(26)	0.4627(41)	0.6716(24)	15.2(16)
N(110)	0.7186(16)	0.4677(24)	0.6220(14)	9.2(8)
C(111)	0.7298(22)	0.4807(35)	0.5721(18)	9.9(13)
C(112)	0.7611(19)	0.3989(33)	0.5577(16)	10.0(11)
O(113)	0.7277(11)	0.3151(20)	0.5559(9)	8.3(6)
C(114)	0.7573(21)	0.2276(36)	0.5483(18)	9.6(12)
C(115)	0.7250(18)	0.1441(28)	0.5582(15)	7.7(10)
O(116)	0.7270(9)	0.1506(15)	0.6150(8)	6.9(5)
C(117)	0.7136(17)	0.0599(27)	0.6351(14)	7.8(9)
C(118)	0.6499(17)	0.0237(28)	0.6100(14)	7.8(9)
C(119)	0.5465(18)	0.0930(28)	0.5779(16)	9.4(10)
C(120)	0.5495(20)	0.1347(34)	0.5232(18)	11.6(12)
O(121)	0.5624(13)	0.2400(21)	0.5323(11)	9.0(7)
C(122)	0.5170(22)	0.2992(36)	0.5446(19)	12.5(14)
C(123)	0.5356(17)	0.4039(28)	0.5468(14)	8.8(9)
O(124)	0.5881(13)	0.4238(21)	0.5892(11)	10.4(7)
C(125)	0.6142(28)	0.5203(44)	0.5952(22)	14.2(16)
C(126)	0.6690(27)	0.5316(38)	0.6289(21)	13.4(15)

Table 8. (Continued)

Positional and thermal parameters for the second crypt

	x	y	z	B
N(201)	0.6393(16)	0.3287(27)	0.3723(14)	10.8(9)
C(202)	0.6877(23)	0.3230(36)	0.3491(19)	12(1)
C(203)	0.6834(27)	0.3905(42)	0.3088(25)	15(2)
O(204)	0.6323(16)	0.3958(25)	0.2572(14)	13.8(9)
C(205)	0.6375(22)	0.3237(35)	0.2169(20)	12(1)
C(206)	0.5836(27)	0.3316(37)	0.1777(21)	13(1)
O(207)	0.5366(21)	0.2890(29)	0.1940(16)	17(1)
C(208)	0.4652(38)	0.2839(55)	0.1526(30)	20(2)
C(209)	0.4262(50)	0.3642(89)	0.1543(43)	28(4)
N(210)	0.4335(29)	0.4466(52)	0.1871(24)	18(2)
C(211)	0.4528(35)	0.5520(62)	0.1852(31)	20(2)
C(212)	0.4652(72)	0.569(11)	0.2380(68)	38(8)
O(213)	0.5142(19)	0.5571(29)	0.2652(18)	16(1)
C(214)	0.5175(22)	0.6048(34)	0.3097(21)	11(1)
C(215)	0.5612(23)	0.5790(38)	0.3566(20)	13(1)
O(216)	0.5505(13)	0.4818(21)	0.3663(11)	10.7(7)
C(217)	0.5906(24)	0.4515(35)	0.4175(20)	13(1)
C(218)	0.6450(23)	0.4133(36)	0.4138(19)	13(1)
C(219)	0.6232(23)	0.2358(39)	0.3994(21)	13(1)
C(220)	0.5872(33)	0.1727(47)	0.3612(27)	18(2)
O(221)	0.5265(15)	0.2052(22)	0.3307(11)	12.0(8)
C(222)	0.4756(29)	0.2090(41)	0.3359(23)	15(2)
C(223)	0.4266(33)	0.2429(56)	0.3189(30)	18(2)
O(224)	0.4168(22)	0.3193(37)	0.2889(22)	20(2)
C(225)	0.3742(36)	0.3351(60)	0.2292(35)	21(3)
C(226)	0.3866(36)	0.4153(58)	0.2034(29)	19(2)

Table 8. (Continued)

Positional and thermal parameters for the third crypt[c]

	x	y	z	B
N(301)	0.5912(19)	0.9531(29)	0.2287(16)	12(1)
C(302)	0.5654(25)	0.9192(40)	0.2671(21)	12(2)
C(303)	0.4978(27)	0.9655(44)	0.2184(22)	14(2)
O(304)	0.4822(16)	0.8642(25)	0.1816(13)	12(1)
C(305)	0.4581(32)	0.9486(49)	0.1420(25)	17(2)
C(306)	0.4244(24)	0.8824(37)	0.1043(20)	11(1)
O(307)	0.4695(17)	0.8624(27)	0.0658(14)	13(1)
C(308)	0.4409(27)	0.7681(42)	0.0576(22)	14(2)
C(309)	0.4797(26)	0.7426(40)	0.0036(23)	13(2)
N(310)	0.5493(19)	0.6942(28)	0.0310(15)	11(1)
C(311)	0.5997(24)	0.7262(36)	0.0157(20)	11(1)
C(312)	0.5872(27)	0.8393(39)	0.0014(22)	13(2)
O(313)	0.6304(18)	0.8426(28)	0.0541(15)	14(1)
C(314)	0.6559(25)	0.9532(39)	0.0603(20)	12(2)
C(315)	0.6097(26)	0.0060(42)	0.0657(21)	13(2)
O(316)	0.5990(18)	0.0174(28)	0.1238(14)	14(1)
C(317)	0.6508(24)	0.0092(38)	0.1780(19)	11(2)
C(318)	0.6294(22)	0.0299(35)	0.2191(18)	10(1)
C(319)	0.6555(33)	0.8977(52)	0.2578(27)	18(2)
C(320)	0.6207(28)	0.7667(45)	0.2542(23)	14(2)
O(321)	0.6721(20)	0.7624(32)	0.2240(17)	16(1)
C(322)	0.6907(31)	0.6851(45)	0.2042(25)	16(2)
C(323)	0.6526(31)	0.6332(46)	0.1660(29)	16(2)
O(324)	0.5958(18)	0.6157(29)	0.1411(15)	14(2)
C(325)	0.5819(26)	0.5787(40)	0.0813(21)	13(2)
C(326)	0.5282(26)	0.6252(40)	0.0589(21)	12(2)

[c]Estimated standard deviations in parameters from FBLS[78] are in parentheses.

Table 9. Interatomic distances and angles[a] for the Sb_7^{3-} anion

Distances (Å)

	Sb(2)	Sb(3)	Sb(4)	Sb(5)	Sb(6)	Sb(7)
Sb(1)	2.711(16)	2.776(43)	4.339(69)	4.190(19)	4.432(24)	4.365(14)
Sb(2)		4.209(12)	4.432(52)	4.434(25)	2.838(46)	2.880(12)
Sb(3)			2.755(15)	2.824(22)	4.159(14)	4.211(34)
Sb(4)				4.313(5)	2.708(8)	4.448(12)
Sb(5)					4.409(4)	2.693(6)
Sb(6)						2.856(5)

Bonded Angles

Atom 1	Atom 2 (Vertex)	Atom 3	Degrees	Atom 1	Atom 2 (Vertex)	Atom 3	Degrees
Sb(1)	Sb(3)	Sb(4)	103.4(8)	Sb(5)	Sb(7)	Sb(2)	105.4(6)
Sb(4)	Sb(3)	Sb(5)	101.3(1)	Sb(5)	Sb(7)	Sb(6)	105.2(2)
Sb(1)	Sb(3)	Sb(5)	96.9(2)	Sb(1)	Sb(2)	Sb(6)	106.0(1)
Sb(2)	Sb(6)	Sb(7)	60.8(3)	Sb(1)	Sb(2)	Sb(7)	102.6(4)
Sb(6)	Sb(7)	Sb(2)	59.3(9)	Sb(4)	Sb(6)	Sb(2)	106.1(7)
Sb(7)	Sb(2)	Sb(6)	59.9(7)	Sb(4)	Sb(6)	Sb(7)	106.1(2)
Sb(3)	Sb(5)	Sb(7)	99.5(5)				
Sb(3)	Sb(1)	Sb(2)	100.2(3)				
Sb(3)	Sb(4)	Sb(6)	99.2(6)				

[a]Estimated standard deviations form ORFFE[65] are in parentheses.

Table 10. Interatomic distances and angles[a] for the first cryptated sodium cation

Bonded Distances			Nonbonded Distances		
Atom 1	Atom 2	Distance (Å)	Atom 1	Atom 2	Distance (Å)
N(101)	C(102)	1.52(5)	N(101)	N(110)	5.76(4)
C(102)	C(103)	1.52(5)	N(101)	O(104)	2.97(4)
C(103)	O(104)	1.40(4)	N(101)	O(116)	2.96(4)
O(104)	C(105)	1.47(5)	N(101)	O(121)	2.90(4)
C(105)	C(106)	1.40(5)	N(110)	O(107)	2.87(4)
C(106)	O(107)	1.44(5)	N(110)	O(113)	2.74(4)
O(107)	C(108)	1.47(5)	N(110)	O(124)	2.95(5)
C(108)	C(109)	1.29(6)	O(104)	O(107)	2.72(4)
C(109)	N(110)	1.44(6)	O(104)	O(113)	5.06(5)
N(110)	C(111)	1.38(5)	O(104)	O(116)	3.94(4)
N(110)	C(126)	1.51(5)	O(104)	O(121)	4.11(5)
C(111)	C(112)	1.45(5)	O(104)	O(124)	3.62(4)
C(112)	O(113)	1.39(4)	O(107)	O(113)	3.91(3)
O(113)	C(114)	1.43(5)	O(107)	O(116)	3.47(3)
C(114)	C(115)	1.44(5)	O(107)	O(121)	5.22(9)
C(115)	O(116)	1.43(4)	O(107)	O(124)	4.06(7)
O(116)	C(117)	1.42(4)	O(113)	O(116)	2.72(3)
C(117)	C(118)	1.50(5)	O(113)	O(121)	3.85(4)
C(118)	N(101)	1.49(4)	O(113)	O(124)	3.91(4)
N(101)	C(119)	1.43(5)	O(116)	O(121)	3.94(6)
C(119)	C(120)	1.52(5)	O(116)	O(124)	4.87(4)
C(120)	O(121)	1.49(5)	O(121)	O(124)	2.88(4)
O(121)	C(122)	1.45(5)	Na(1)	C(102)	3.64(5)
C(122)	C(123)	1.50(5)	Na(1)	C(103)	3.41(5)
C(123)	O(124)	1.37(4)	Na(1)	C(105)	3.36(5)
O(124)	C(125)	1.45(6)	Na(1)	C(106)	3.50(6)
C(125)	C(126)	1.30(6)	Na(1)	C(108)	3.53(7)
			Na(1)	C(109)	3.46(6)
Na(1)	N(101)	2.94(3)	Na(1)	C(111)	3.51(5)
Na(1)	N(110)	2.83(3)	Na(1)	C(112)	3.51(5)
Na(1)	O(104)	2.56(3)	Na(1)	C(114)	3.34(6)
Na(1)	O(107)	2.55(5)	Na(1)	C(115)	3.10(4)
Na(1)	O(113)	2.55(4)	Na(1)	C(117)	3.30(4)
Na(1)	O(116)	2.40(2)	Na(1)	C(118)	3.61(4)
Na(1)	O(121)	2.71(5)	Na(1)	C(119)	3.69(5)
Na(1)	O(124)	2.54(3)	Na(1)	C(120)	3.61(6)
			Na(1)	C(122)	3.32(7)
			Na(1)	C(123)	3.37(5)
			Na(1)	C(125)	3.43(6)
			Na(1)	C(126)	3.41(5)

[a]Estimated standard deviations from ORFFE[65] are in parentheses.

Table 10. (Continued)

Bonded Distances

Atom 1	Atom 2	Distance (Å)

Nonbonded Distances

Atom 1	Atom 2	Distance (Å)
C(119)	Sb(4)	3.88(4)
C(120)	Sb(4)	4.49(5)
C(122)	Sb(3)	4.26(6)
C(122)	Sb(4)	4.48(5)

Bonded Angles

Atom 1	Atom 2 (Vertex)	Atom 3	Degrees
N(101)	C(102)	C(103)	109(3)
C(102)	C(103)	O(104)	112(4)
C(103)	O(104)	C(105)	117(3)
O(104)	C(105)	C(106)	114(4)
C(105)	C(106)	O(107)	106(4)
C(106)	O(107)	C(108)	105(3)
O(107)	C(108)	C(109)	114(5)
C(108)	C(109)	N(110)	126(6)
C(109)	N(110)	C(111)	117(4)
C(109)	N(110)	C(126)	113(4)
C(111)	N(110)	C(126)	114(4)
N(110)	C(111)	C(112)	113(4)
C(111)	C(112)	O(113)	110(4)
C(112)	O(113)	C(114)	115(3)
O(113)	C(114)	C(115)	110(4)
C(114)	C(115)	O(116)	106(3)
C(115)	O(116)	C(117)	111(2)
O(116)	C(117)	C(118)	116(3)
C(117)	C(118)	N(101)	111(3)
C(118)	N(101)	C(102)	110(3)
C(118)	N(101)	C(119)	116(3)
C(119)	N(101)	C(102)	111(3)
N(101)	C(119)	C(120)	110(4)
C(119)	C(120)	O(121)	107(3)
C(120)	O(121)	C(122)	117(3)
O(121)	C(122)	C(123)	109(4)
C(122)	C(123)	O(124)	113(3)
C(123)	O(124)	C(125)	120(4)
O(124)	C(125)	C(126)	118(5)
C(125)	C(126)	N(110)	119(5)
N(101)	Na(1)	N(110)	178.2(10)
N(101)	Na(1)	O(104)	65.0(9)
N(101)	Na(1)	O(116)	66.4(9)
N(101)	Na(1)	O(121)	61.6(9)
N(110)	Na(1)	O(107)	64.4(10)
N(110)	Na(1)	O(113)	61.0(10)
N(110)	Na(1)	O(124)	66.4(11)
O(104)	Na(1)	O(107)	64.3(9)
O(104)	Na(1)	O(113)	163.1(10)
O(104)	Na(1)	O(116)	105.0(10)
O(104)	Na(1)	O(121)	102.3(11)
O(104)	Na(1)	O(124)	90.6(11)
O(107)	Na(1)	O(113)	100.1(10)
O(107)	Na(1)	O(116)	89.2(11)
O(107)	Na(1)	O(121)	165.4(11)
O(107)	Na(1)	O(124)	106.0(13)
O(113)	Na(1)	O(116)	66.6(9)
O(113)	Na(1)	O(121)	93.8(9)
O(113)	Na(1)	O(124)	100.4(10)
O(116)	Na(1)	O(121)	100.6(10)
O(116)	Na(1)	O(124)	161.9(9)
O(121)	Na(1)	O(124)	66.6(11)

Table 11. Interatomic distances and angles[a] for the second cryptated sodium cation

Bonded Distances			Nonbonded Distances		
Atom 1	Atom 2	Distance (Å)	Atom 1	Atom 2	Distance (Å)
N(201)	C(202)	1.43(5)	N(201)	N(210)	5.77(11)
C(202)	C(203)	1.36(6)	N(201)	O(204)	3.02(5)
C(203)	O(204)	1.46(6)	N(201)	O(216)	2.93(5)
O(204)	C(205)	1.46(5)	N(201)	O(221)	3.02(5)
C(205)	C(206)	1.33(6)	N(210)	O(207)	3.20(8)
C(206)	O(207)	1.41(6)	N(210)	O(213)	2.72(9)
O(207)	C(208)	1.66(8)	N(210)	O(224)	3.24(8)
C(208)	C(209)	1.44(11)	O(204)	O(207)	2.73(6)
C(209)	N(210)	1.39(11)	O(204)	O(213)	3.59(5)
N(210)	C(211)	1.53(8)	O(204)	O(216)	4.01(5)
N(210)	C(226)	1.35(8)	O(204)	O(221)	4.40(5)
C(211)	C(212)	1.30(15)	O(204)	O(224)	5.43(6)
C(212)	O(213)	1.14(15)	O(207)	O(213)	4.22(6)
O(213)	C(214)	1.29(5)	O(207)	O(216)	5.04(5)
C(214)	C(215)	1.34(6)	O(207)	O(221)	3.73(5)
C(215)	O(216)	1.40(5)	O(207)	O(224)	4.25(8)
O(216)	C(217)	1.40(5)	O(213)	O(216)	2.64(5)
C(217)	C(218)	1.40(6)	O(213)	O(221)	5.11(5)
C(218)	N(201)	1.55(5)	O(213)	O(224)	4.14(6)
N(201)	C(219)	1.55(5)	O(216)	O(221)	3.92(4)
C(219)	C(220)	1.37(7)	O(216)	O(224)	2.83(7)
C(220)	O(221)	1.45(7)	O(221)	O(224)	2.90(6)
O(221)	C(222)	1.52(6)			
C(222)	C(223)	1.31(8)	Na(2)	C(202)	3.56(6)
C(223)	O(224)	1.28(7)	Na(2)	C(203)	3.42(6)
O(224)	C(225)	1.54(8)	Na(2)	C(205)	3.50(6)
C(225)	C(226)	1.36(9)	Na(2)	C(206)	3.36(6)
			Na(2)	C(208)	3.42(8)
Na(2)	N(201)	2.85(6)	Na(2)	C(209)	3.42(12)
Na(2)	N(210)	2.95(7)	Na(2)	C(211)	3.59(8)
Na(2)	O(204)	2.69(4)	Na(2)	C(212)	3.16(16)
Na(2)	O(207)	2.60(4)	Na(2)	C(214)	3.32(5)
Na(2)	O(213)	2.61(4)	Na(2)	C(215)	3.34(5)
Na(2)	O(216)	2.50(3)	Na(2)	C(217)	3.40(6)
Na(2)	O(221)	2.59(3)	Na(2)	C(218)	3.55(8)
Na(2)	O(224)	2.78(5)	Na(2)	C(219)	3.56(7)

[a]Estimated standard deviations from ORFFE[65] are in parentheses.

Table 11. (Continued)

Bonded Distances			Nonbonded Distances		
Atom 1	Atom 2	Distance (Å)	Atom 1	Atom 2	Distance (Å)
			Na(2)	C(220)	3.38(7)
			Na(2)	C(222)	3.38(6)
			Na(2)	C(223)	3.33(7)
			Na(2)	C(225)	3.50(9)
			Na(2)	C(226)	3.38(9)

Bonded Angles

Atom 1	Atom 2 (Vertex)	Atom 3	Degrees	Atom 1	Atom 2 (Vertex)	Atom 3	Degrees
N(201)	C(202)	C(203)	113(5)	C(223)	O(224)	C(225)	130(6)
C(202)	C(203)	O(204)	123(5)	O(224)	C(225)	C(226)	115(7)
C(203)	O(204)	C(205)	112(4)	C(225)	C(226)	N(210)	136(8)
O(204)	C(205)	C(206)	101(4)				
C(205)	C(206)	O(207)	112(5)	N(201)	Na(2)	N(210)	168(2)
C(206)	O(207)	C(208)	123(5)	N(201)	Na(2)	O(204)	66(1)
O(207)	C(208)	C(209)	118(7)	N(201)	Na(2)	O(216)	66(1)
C(208)	C(209)	N(210)	134(10)	N(201)	Na(2)	O(221)	67(1)
C(209)	N(210)	C(211)	138(8)	N(210)	Na(2)	O(207)	70(2)
C(209)	N(210)	C(226)	88(7)	N(210)	Na(2)	O(213)	58(2)
C(211)	N(210)	C(226)	126(7)	N(210)	Na(2)	O(224)	69(2)
N(210)	C(211)	C(212)	96(9)	O(204)	Na(2)	O(207)	62(2)
C(211)	C(212)	O(213)	116(15)	O(204)	Na(2)	O(213)	85(1)
C(212)	O(213)	C(214)	103(10)	O(204)	Na(2)	O(216)	101(1)
O(213)	C(214)	C(215)	117(5)	O(204)	Na(2)	O(221)	113(1)
C(214)	C(215)	O(216)	106(4)	O(204)	Na(2)	O(224)	166(1)
C(215)	O(216)	C(217)	110(4)	O(207)	Na(2)	O(213)	108(1)
O(216)	C(217)	C(218)	113(4)	O(207)	Na(2)	O(216)	162(1)
C(217)	C(218)	N(201)	116(4)	O(207)	Na(2)	O(221)	92(1)
C(218)	N(201)	C(202)	114(4)	O(207)	Na(2)	O(224)	104(2)
C(218)	N(201)	C(219)	107(3)	O(213)	Na(2)	O(216)	62(1)
C(219)	N(201)	C(202)	118(4)	O(213)	Na(2)	O(221)	158(1)
N(201)	C(219)	C(220)	113(5)	O(213)	Na(2)	O(224)	100(1)
C(219)	C(220)	O(221)	117(6)	O(216)	Na(2)	O(221)	101(1)
C(220)	O(221)	C(222)	123(4)	O(216)	Na(2)	O(224)	93(2)
O(221)	C(222)	C(223)	109(5)	O(221)	Na(2)	O(224)	65(1)
C(222)	C(223)	O(224)	132(8)				

Table 12. Interatomic distances and angles[a] for the third cryptated sodium cation

Bonded Distances			Nonbonded Distances		
Atom 1	Atom 2	Distance (Å)	Atom 1	Atom 2	Distance (Å)
N(301)	C(302)	1.38(8)	N(301)	N(310)	5.98(7)
C(302)	C(303)	1.78(8)	N(301)	O(304)	2.73(7)
C(303)	O(304)	1.65(7)	N(301)	O(116)	2.86(7)
O(304)	C(305)	1.52(8)	N(301)	O(121)	3.26(7)
C(305)	C(306)	1.37(9)	N(310)	O(307)	3.26(6)
C(306)	O(307)	1.67(7)	N(310)	O(313)	2.72(6)
O(307)	C(308)	1.45(7)	N(310)	O(324)	2.87(6)
C(308)	C(309)	1.90(9)	O(304)	O(307)	2.86(5)
C(309)	N(310)	1.68(7)	O(304)	O(313)	5.44(5)
N(310)	C(311)	1.42(7)	O(304)	O(316)	4.07(5)
N(310)	C(326)	1.37(7)	O(304)	O(321)	4.43(6)
C(311)	C(312)	1.61(8)	O(304)	O(324)	4.64(5)
C(312)	O(313)	1.39(7)	O(307)	O(313)	3.86(6)
O(313)	C(314)	1.63(7)	O(307)	O(316)	3.61(6)
C(314)	C(315)	1.34(8)	O(307)	O(321)	5.31(6)
C(315)	O(316)	1.58(7)	O(307)	O(324)	4.50(6)
O(316)	C(317)	1.52(7)	O(313)	O(316)	3.21(6)
C(317)	C(318)	1.32(8)	O(313)	O(321)	4.25(6)
C(318)	N(301)	1.45(8)	O(313)	O(324)	4.05(6)
N(301)	C(319)	1.63(9)	O(316)	O(321)	4.36(6)
C(319)	C(320)	1.97(10)	O(316)	O(324)	5.56(6)
C(320)	O(321)	1.61(8)	O(321)	O(324)	3.05(6)
O(321)	C(322)	1.31(8)			
C(322)	C(323)	1.30(9)	Na(3)	C(302)	3.63(6)
C(323)	O(324)	1.29(8)	Na(3)	C(303)	3.71(6)
O(324)	C(325)	1.53(7)	Na(3)	C(305)	3.27(6)
C(325)	C(326)	1.36(8)	Na(3)	C(306)	3.47(6)
			Na(3)	C(308)	3.27(6)
Na(3)	N(301)	2.84(6)	Na(3)	C(309)	3.61(6)
Na(3)	N(310)	3.14(5)	Na(3)	C(311)	3.56(6)
Na(3)	O(304)	2.84(4)	Na(3)	C(312)	3.51(6)
Na(3)	O(307)	2.62(4)	Na(3)	C(314)	3.51(6)
Na(3)	O(313)	2.76(4)	Na(3)	C(315)	3.27(6)
Na(3)	O(316)	2.70(4)	Na(3)	C(317)	3.02(6)
Na(3)	O(321)	2.74(5)	Na(3)	C(318)	3.46(6)
Na(3)	O(324)	2.96(4)	Na(3)	C(319)	3.19(7)

[a]Estimated standard deviations from FBOND[78] are in parentheses.

Table 12. (Continued)

Bonded Distances			Nonbonded Distances		
Atom 1	Atom 2	Distance (Å)	Atom 1	Atom 2	Distance (Å)
			Na(3)	C(320)	2.96(7)
			Na(3)	C(322)	3.31(7)
			Na(3)	C(323)	3.17(7)
			Na(3)	C(325)	3.73(6)
			Na(3)	C(326)	3.40(6)

Bonded Angles

Atom 1	Atom 2 (Vertex)	Atom 3	Degrees
N(301)	C(302)	C(303)	83(4)
C(302)	C(303)	O(304)	94(4)
C(303)	O(304)	C(305)	72(4)
O(304)	C(305)	C(306)	88(5)
C(305)	C(306)	O(307)	101(4)
C(306)	O(307)	C(308)	83(4)
O(307)	C(308)	C(309)	88(4)
C(308)	C(309)	N(310)	113(4)
C(309)	N(310)	C(311)	122(4)
C(309)	N(310)	C(326)	92(4)
C(311)	N(310)	C(326)	145(5)
N(310)	C(311)	C(312)	105(4)
C(311)	C(312)	O(313)	78(4)
C(312)	O(313)	C(314)	105(4)
O(313)	C(314)	C(315)	104(5)
C(314)	C(315)	O(316)	121(5)
C(315)	O(316)	C(317)	121(4)
O(316)	C(317)	C(318)	108(5)
C(317)	C(318)	N(301)	112(5)
C(318)	N(301)	C(302)	142(5)
C(318)	N(301)	C(319)	84(4)
C(319)	N(301)	C(302)	94(5)
N(301)	C(319)	C(320)	96(5)
C(319)	C(320)	O(321)	73(5)
C(320)	O(321)	O(322)	127(5)
O(321)	C(322)	C(323)	120(6)
C(322)	C(323)	O(324)	145(7)
C(323)	O(324)	C(325)	114(5)
O(324)	C(325)	C(326)	98(5)
C(325)	C(326)	N(310)	96(5)

final least squares cycle. Interatomic distances and angles for the Sb_7^{3-} anion are given in Table 9 while distances and angles for the first and second crypts are given in Table 10 and 11, respectively. The program FBOND[78] was used to obtain bond distances and angles for the third crypt, and these are listed in Table 12.

Discussion

The packing of three cryptated sodium cations per Sb_7^{3-} cluster anion is shown in a projection of the unit cell on the (010) plane in Figure 3.[1] For clarity the cryptated sodium cations are represented simply as a N-Na-N linkage, and the three independent cryptated cations are identified by numerals.

The anion

The Sb_7^{3-} anion is not only the first well-characterized homopolyatomic anion for a metallic element, but it is also apparently one of only a few rigorously established examples of a seven atom polyhedron.[6] The geometry of the anion is shown in Figure 4. The seven antimony atoms are clustered in an end-capped trigonal prism having approximate C_{3v} symmetry.

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[1]The projection of the unit cell on the (010) plane and plots of the Sb_7^{3-} anion and cryptated sodium cations that follow were plotted using the thermal ellipsoid plotting program ORTEP.[64]

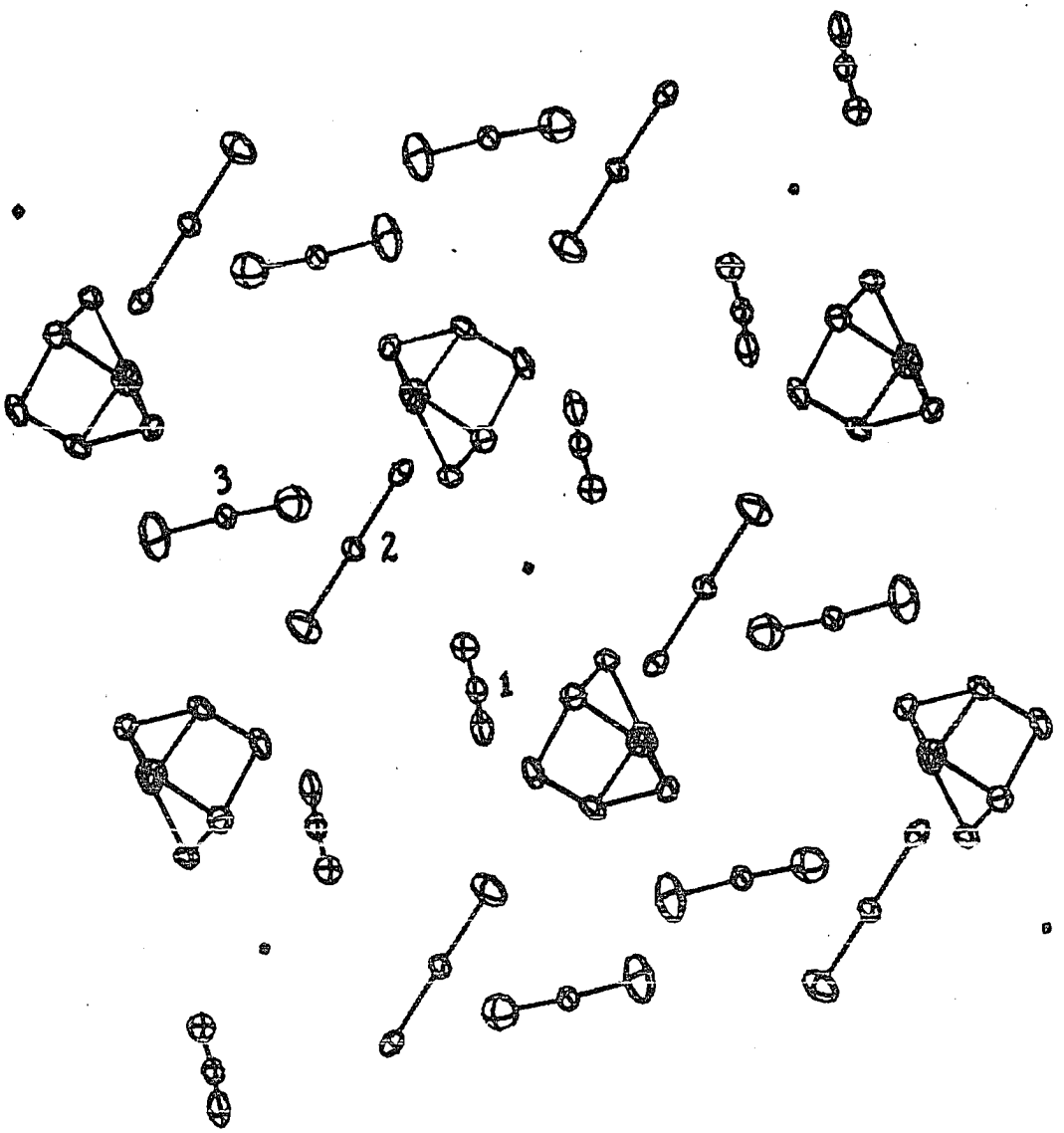

Figure 3. Projection of the structure of $(NaC^{+})_3Sb_7^{3-}$ on the (010) plane.

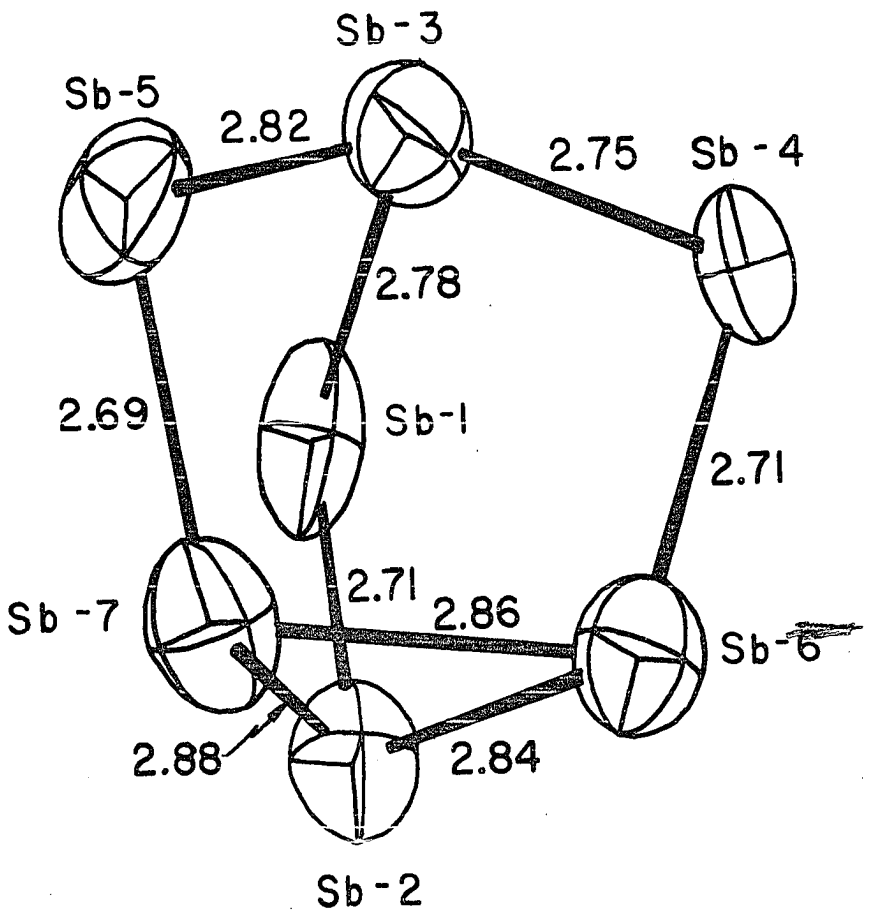

Figure 4. Geometry of the Sb_7^{3-} anion in $(NaC^+)_3Sb_7^{3-}$.

Least squares planes have been calculated[1] for the three four-atom faces: 1,2,6,4; 1,2,7,5; and 5,7,6,4. The perpendicular distance of each of the four atoms from the respective plane is given in Table 13. The angle between the normals to the planes determined by atoms 4,5,1 and 6,7,2 is 0.5(7)°.[65]

Table 13. Deviations from least squares planes

Atom	Distance, Å	Atom	Distance, Å	Atom	Distance, Å
Sb(4)	0.0042	Sb(1)	-0.0189	Sb(1)	0.0287
Sb(5)	-0.0042	Sb(2)	0.0276	Sb(2)	-0.0439
Sb(6)	-0.0063	Sb(5)	0.0193	Sb(4)	-0.0288
Sb(7)	0.0063	Sb(7)	-0.0280	Sb(6)	0.0440

A few years ago Muetterties and Wright reviewed the literature concerning molecular polyhedra of high coordination number.[80] Important polyhedra for the seven-atom family included the capped octahedron (C_{3v}), the pentagonal bipyramid (D_{5h}) and the face-capped trigonal prism (C_{2v}). A fourth basic geometry for seven-coordination which has been rigorously

[1]Least squares planes were calculated using the local program LSP2 written by Wayne Rohrbaugh.

established for complexes, metal clusters and polymeric oxides is the tetragonal base-trigonal base (C_s) polyhedron. It is important to note that these polyhedra have been proposed for ML_7 complexes rather than for a homopolyatomic noncentered cluster as is under consideration here. The C_{3v}, D_{5h} and C_{2v} idealized polyhedra represent limiting symmetry forms as opposed to energetically favored or experimentally established geometries. Conformational interconversions require only slight bending modes, and the energy differences between these idealized geometries should be small with respect to intermolecular forces generated by ordering or by solvation phenomena in the solid, liquid or solution states.[81] Gillespie, among others, has evaluated the relative stabilities of the MX_7 models simply by considering repulsive forces generated from interactions of like ligands constrained to a spherical surface.[82] The analysis has limited predictive value, however, as the energy differences are very small, attractive forces are ignored and calculations are limited to spherical models.

Recently Muetterties and Guggenberger have emphasized that there is a paucity of available structural data for the seven atom family.[83] While there are no good observed C_{3v}-type structures, in $Rb_5Zr_4F_{21}$ there are two independent ZrF_7 polyhedra with the same structure which is midway between the C_{3v} and D_{5h} geometries.[84] Among several complexes which

have been reported which are close to the pentagonal bipyramidal structure are ZrF_7^{3-},[85] ReF_7,[86] IF_7[87] and $V(CN)_7^{4-}$.[88] Two compounds $Na_5Zr_2F_{13}$[89] and K_2NbF_7[90] have been reported which adopt the C_{2v} structure. The structure found for the Sb_7^{3-} anion does not approach any of these idealized polyhedra. It is similar, however, to the structure of P_4S_3. Here phosphorus atoms form a tetrahedron and sulfur atoms bridge along three adjacent edges of the tetrahedron.[8] If outer shell p electrons are considered, the two clusters are isoelectronic. Using a Lewis electron-pair scheme one has a cluster with nine electron-pair bonds and a lone pair on each bridging atom.

The cations

For each of the three independent cryptated cations the sodium ion occupies the central cavity of the macrobicycle. The coordination polyhedron about sodium is bicapped trigonal antiprismatic. The geometry of the first and second cryptated sodium cations is illustrated in Figures 5 and 6, respectively. The bond distances indicated in the figures were calculated prior to the final lattice constant refinement. The numbering system for the third crypt which is not illustrated parallels that of the other two. Moras, Metz and Weiss noted that as the size of the alkali cation decreases from cesium to sodium, the oxygen coordination changes from approximately trigonal prismatic in the case

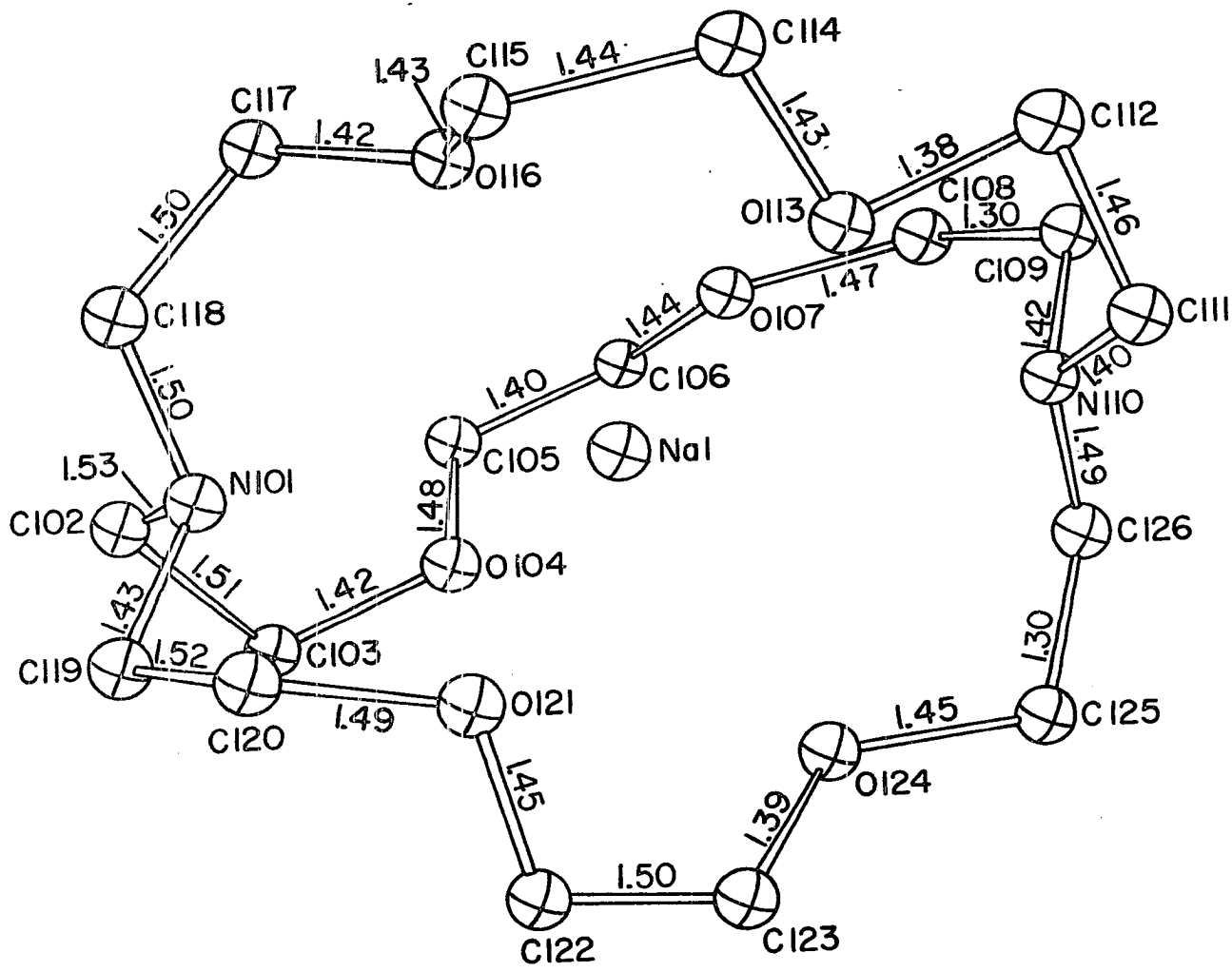

Figure 5. Geometry of the first cryptated sodium cation in $(NaC^{+})_3Sb_7^{3-}$.

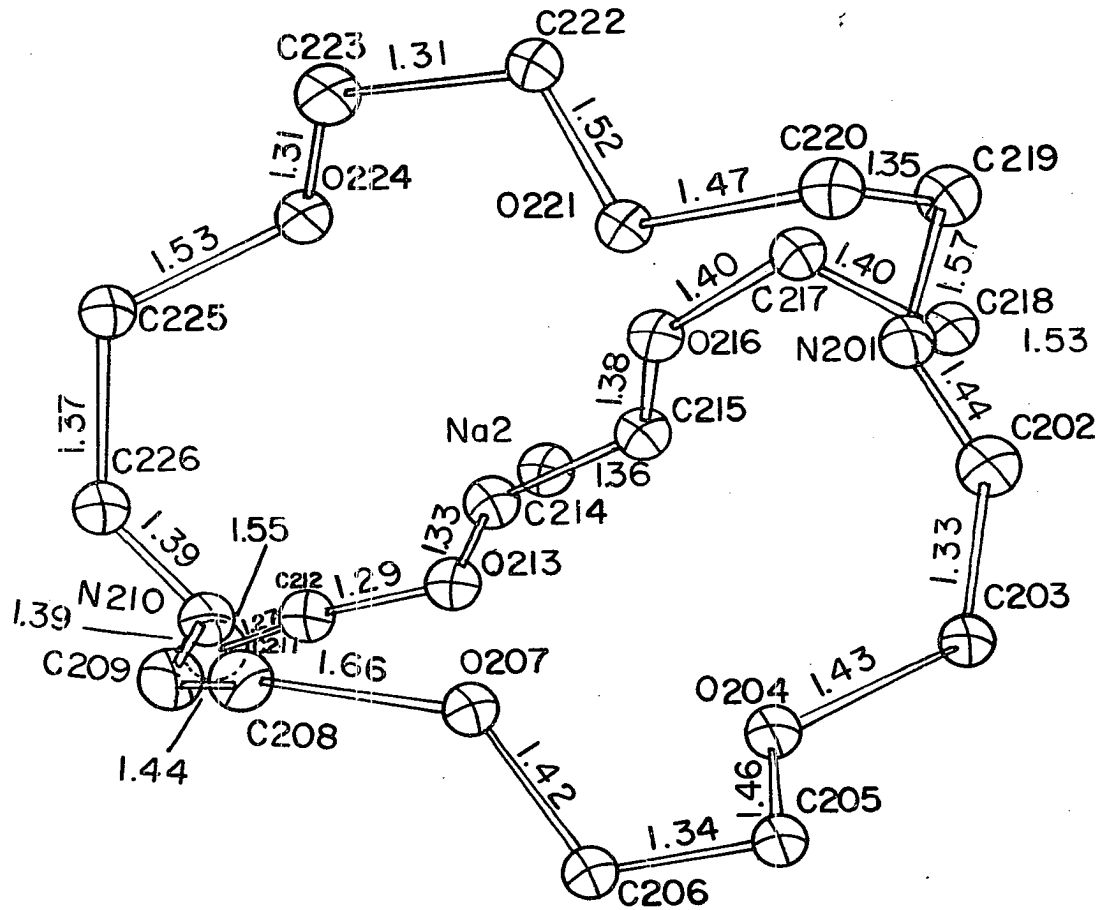

Figure 6. Geometry of the second cryptated sodium cation in $(NaC^{+})_3Sb_7^{3-}$.

of CsC^+I^- and RbC^+I^-[91] and KC^+I^-[92] to very nearly trigonal antiprismatic in the case of NaC^+I^-.[42] The conformational flexibility of 2,2,2-crypt was also noted by Dye et al. who likewise found trigonal antiprismatic coordination about sodium in NaC^+Na^-[38] and confirmed the ability of the strands to twist. While free 2,2,2-crypt exists in solution as an equilibrium mixture of three conformations having the bridgehead nitrogens either out-out, in-out, or in-in,[93] all crystal structures so far determined for cryptated metal ions logically have contained crypt in the in-in conformation.[41] The N(101)-N(110) distance of 5.76Å found here compares with 6.87Å in free crypt.[94]

A convenient way to consider the atom placements in the present structure is by comparison with the known structures of NaC^+Na^-[38] and NaC^+I^-.[42] The former compound crystallizes in the space group R32 while the latter one crystallizes in the space group P31c. Although P31c does not have the two-fold rotation axes of R32, both space groups have three-fold symmetry. Interatomic distances and angles found for the two compounds are compared with averages for the first crypt in Table 14.

The refinement reported here is more satisfactory for the first crypt than for the other two. That of the second crypt is reasonably satisfactory with the exception of the atoms near the N(210) bridgehead. While the refinement of the third crypt is much less satisfactory than for the other

Table 14. Interatomic distances and angles for cryptated sodium cations

Bonded Distances (Å)	NaC^+Na^- [a]	NaC^+I^- [b]	Crypt 1 [b]
Na-N	2.72(1)	2.75(2)	2.88(8)
Na-O	2.57(1)	2.58(2)	2.55(10)
N-C1	1.48(2)	1.47(2)	1.46(5)
C1-C2	1.29(2)	1.44(3)	1.43(11)
C2-O	1.41(1)	1.44(2)	1.44(4)
O-C3	1.40(2)	1.40(5)	1.43(3)
C3-C3(2)	1.31(3)	1.40(4)	1.45(5)
Nonbonded Distances (Å)			
Na-C1	3.45(2)	3.53(2)	3.55(11)
Na-C2	3.46(2)	3.46(2)	3.46(11)
Na-C3	3.34(3)	3.39(2)	3.33(13)
O-O(2)	2.86(2)	2.79(2)	2.77(9)
O-O(3)	4.09(2)	4.07(2)	3.98(9)
$O\text{-}O(3^12)$	5.13(2)	5.12(2)	5.05(17)
$O\text{-}O(3^22)$	3.38(2)	3.53(2)	3.65(19)
N-N(2)	5.44(2)	5.50(2)	5.76
Angles (deg)			
Na-N-C1	107(1)	110(1)	105(4)
C1-N-C1(3)	112(1)	109(1)	113(3)
N-C1-C2	128(1)	113(2)	115(7)
C1-C2-O	120(1)	117(3)	113(4)
C2-O-C3	107(1)	100(2)	114(5)
O-C3-C3(2)	121(2)	116(3)	110(3)
O-Na-O(2)	67.6(3)	65.7(4)	66(1)
O-Na-O(3)	105.4(3)	104.4(4)	102(2)
$O\text{-}Na\text{-}O(3^12)$	171.2(3)	167.0(4)	163(2)
$O\text{-}Na\text{-}O(3^22)$	82.0(3)	86.6(4)	91(2)

[a] Estimated standard deviation of the least significant digit is given in parentheses.

[b] Estimated standard deviation of the average of distances from Table 10 is given in parentheses.

two, inspection of the Na-N, Na-O and Na-C distances in Table 12 assures that the ligand complexing Na(3) is indeed 2,2,2-crypt. The specter of disorder at the bridgeheads N(210), N(301) and N(310) in this structure determination was indicated by multiple carbon positions in the difference maps phased on the basis of only the heavy atom positions. Block least squares refinement of judiciously chosen positions for the atoms of the second and third crypts followed by refinement of the positions and isotropic thermal parameters resulted in considerably larger thermal parameters than obtained in the case of the first crypt. Stout and Jensen have discussed the difficulties arising from disorder which is limited to a given portion of a molecule.[95] They suggest that this type of disorder is indicated by abnormally high thermal parameters for whole atoms located at sites having partial occupancy. The introduction of fractional atom positions for carbon atoms surrounding the N(210) bridgehead of the second crypt and for C(305), C(308), C(309), C(319), C(320), O(321), C(322) and C(323) in the third crypt did result in smaller converged isotropic thermal parameters for those atoms but did not provide a statistically significant[62] improvement in the weighted agreement factor.

It may be noted that in the block least squares refinement of the second crypt more reasonable distances and angles were found than with the full matrix one. For the former

refinement estimated standard deviations for the distances between 0.06 and 0.08Å and for the angles 3.7 and 5.2° were obtained. Apparently to the extent that the block diagonal approach is a rougher approximation than the full-matrix one, errors are distributed over the macrobicycle as a whole. In view of this experience with the second crypt it seemed appropriate to report the block least squares refinement for the third crypt rather than carry out a full-matrix refinement of dubious value. Some consolation is derived from the fact that Dye et al.[38] observed abnormally high thermal parameters for carbon, nitrogen and oxygen atoms of 2,2,2-crypt in $NaC^{+}Na^{-}$ plus unusually short C-C distances of 1.29 and 1.31Å and bond angles at carbon of 128, 120 and 122°. They noted that artificial shortening of interatomic distances and opening of bond angles often occur in structures with high thermal parameters. Among the factors presumably contributing to the difficulties experienced in the refinement reported here is the fact that a thirty per cent decay in the intensities of the standards occurred during data collection. Dye et al. reported observing thermal decomposition during data collection for $NaC^{+}Na^{-}$.[38] However, final atomic parameters obtained from data they collected from a second crystal agreed within 3σ of those determined from the original data. In view of the fact that the goniometer stage conducts heat from the motors driving the four circles of the

diffractometer used here, the $(NaC^{+})_3Sb_7^{3-}$ crystal may have suffered some thermal decomposition during data collection. The effect of instrumental intemperance[63] at the time of data collection is unknown. All of these problems and details are, of course, less significant and interesting than the anion discovered.

Conclusion

The solution of the crystal structure of (cryptated sodium) heptantimonide(-3) provides the first example of a stable solid containing a well-characterized homopolyatomic cluster anion for a metallic element. Only recently Corbett indicated that there is no evidence for solid compounds containing Sb_7^{3-}.[6] The demonstrated success of this synthetic approach as well as the preliminary results of Kummer and Diehl who achieved the isolation of Sn_9^{4-} in salts with sodium complexed with ethylenediamine[36] suggests the desirability of further synthetic effort in this area.

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